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Battelle Pacific Northwest Labs., Richland, WA

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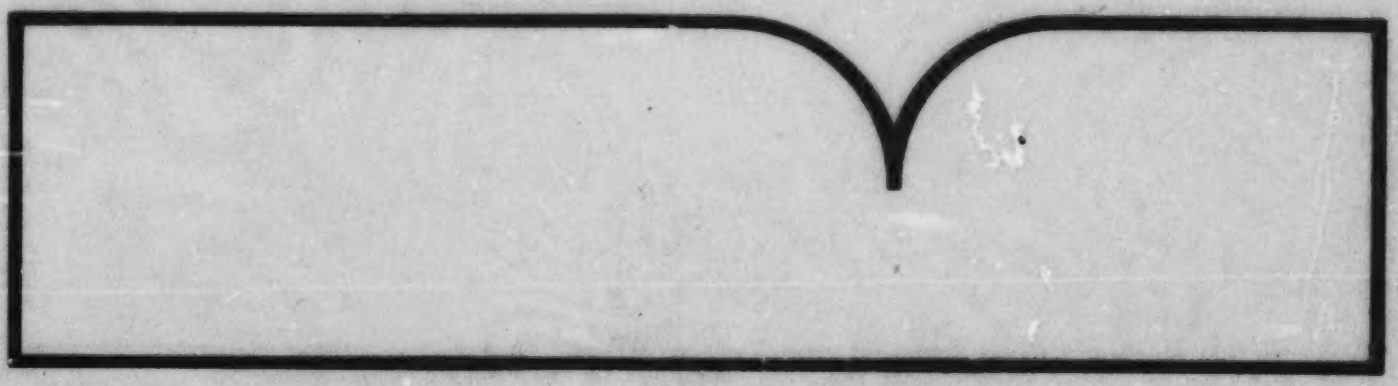

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AN ENVIRONMENTAL STUDY OF ACTIVE AND INACTIVE URANIUM MINES AND THEIR EFFLUENTS

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AN ENVIRONMENTAL STUDY OF ACTIVE AND INACTIVE URANIUM MINES AND THEIR EFFLUENTS

PART I

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AN ENVIRONMENTAL STUDY OF ACTIVE AND INACTIVE URANIUM MINES, MILLS, AND THEIR EFFLUENTS

Pacific Northwest Laboratory
Richland, Washington 99352

ABSTRACT

The purpose of this short-term study has been: 1) to develop broad bases; and 2) to obtain new information relevant to the needs of the Environmental Protection Agency. These data bases are required to provide information for the Clean Air Act and the Uranium Mill Tailings Radiation Control Act. This report, Part I, deals specifically with four tasks associated with the Clean Air Act. They are:

Task 1. Effect of Bulkheading or Filling Underground Mines on Radon Emission

Task 2. Radon Daughter Equilibrium at Various Distances from Exhaust Vents of Underground Uranium mines

Task 3. Incentives for Using Elevated Release Points to Reduce Ground Level Radon Concentrations from Underground Uranium Mines

Task 4. A Study of the Contribution of Multiple Radon Sources in the Ambrosia Lake District, New Mexico, to Concentrations at Selected Locations

Each task is discussed individually in the following sections of this report.

Two open literature publications are being prepared as a part of this study in addition to the data base. They are:

"Bulkheading Effects on Radon Release from the Twilight Uranium Mine," V. W. Thomas

"Relative Contributions of Radon Sources on Ambient Air Concentrations from Mining and Milling of Uranium," J. G. Droppo and J. A. Glissmeyer

Due to the complexity of the data and the number of tasks, each task has its own summary and recommendation section. However, for ease of reading, the Executive Summary lists all detail with respect to the interpretation of data developed in the tasks and their associated recommendations.

EXECUTIVE SUMMARY

TASK 1 - EFFECT OF BULKHEADING OR FILLING UNDERGROUND MINES ON RADON EMISSION

A study of the effects of bulkheading on the radon release from an underground uranium mine was conducted at the Bureau of Mines underground uranium research mine. The research mine is known as the Twilight Mine and is located near Uravan, Colorado. The air flow conditions included exhausting and blowing ventilations with and without bulkheading for six consecutive experimental periods of up to two weeks each. Radon measurements in mine and exhaust air were made for both directions of air flow from bulkhead doors at either end of an internal loop open, closed, and closed but with a major leak. These measurements show that bulkheading produced a major effect on the total radon leaving the mine and that this effect was dependent upon the type of ventilation and the integrity of the bulkheads. During exhausting ventilation, the total radon discharged from the mine decreased by 17% when the bulkhead doors were closed but with a large leak. A further reduction of 35% was observed when the large leak was closed. This latter measurement, however, was made over a very short time period and radon had not built up to its maximum equilibrium concentration. During blowing ventilation, a small (6%) but not significant increase in the total radon leaving the mine was measured when the bulkhead doors were closed but with the large leak. When the large leak was eliminated, a reduction of 35% was observed. Other parameters affecting the total radon leaving the mine, including barometric pressure, must also be taken into consideration when interpreting these measurements. A small amount of air was pumped from the bulkheaded area of the mine to maintain a negative pressure. This air, with its relatively high radon content, could be passed through a sorption trap or discharged at a high elevation to minimize exposure to people in the environs. If this air were passed through absolute sorption traps, it could reduce the radon leaving the mine to 45% and 68%, respectively, of that for exhausting and blowing ventilation with no bulkheading.

Recommendations

While valuable information was obtained from this bulkheading experiment, it is recognized that a large-scale experiment should be conducted in

an operating uranium mine. Battelle scientists have met with the Bureau of Mines and with representatives of a major uranium mining company to discuss both the current experiment and a possible experiment in the future at a large uranium mine.

It seems important to compare the potential of bulkheading in a large active mine with the observations which have been made in this very small-scale study. Whereas the experimental mine had somewhat comparable areas of active and inactive regions, an actual working mine may have worked-out regions with tens of times more volume than the working regions. It can thus be surmised that effective bulkheading of active mines could reduce radon emissions by up to an order of magnitude, or perhaps more. If a study were to be conducted at an active uranium mine, one should be selected where the potential for bulkheading of a substantial region of the mine volume is possible. Discussions with mine operators have indicated their willingness to participate in a study if a well-conceived experiment could be devised. From considerations of active mines, it appears that suitable ones are available and that a study which would include the cooperation of the mine operator, the Bureau of Mines, and PNL could be productively carried out.

TASK 2 - RADON DAUGHTER EQUILIBRIUM AT VARIOUS DISTANCES FROM EXHAUST VENTS OF UNDERGROUND URANIUM MINES

Data obtained in the study indicate that the ratios of radon daughters to radon are relatively low in exhaust air from the underground uranium mine vents. Since working levels are determined by the total concentrations of radon daughters rather than radon itself, this means that the direct exposure to the ventilation exhaust is much less than it would be if the radon daughters more nearly approached equilibrium with radon. The radon daughter equilibrium measurements made downwind from the ventilation exhaust ports showed low ratios indicating that either radon daughters are scavenged by the aerosols that are contained in the plume or that they are scavenged by contact with the earth's surface. In many cases, the data do not suggest a significant build-up in radon daughters with time during transport of the plume at ground level. The working levels were below the proposed indoor EPA standard of 0.015 at all points exceeding 100 meters from the vent.

Recommendations

The measurements which have been made in this study indicate that the ratios of radon daughters to radon are relatively low in exhaust air from the vents. Since working levels are determined by the total concentrations of radon daughters rather than radon itself, this means that the direct exposure to the ventilation exhaust is much less than it would be if the radon daughters more nearly approached equilibrium with radon. Measurements downwind from the ventilation exhaust also show low ratios indicating that either radon daughters are scavenged by the aerosols that are contained in the plume, as is apparently the case for the wet exhaust, or that they are scavenged by contact with the earth's surface. In many cases, the data do not suggest a significant build-up in radon daughters with time during transport of the plume. Difficulties in making the measurements, however, preclude a firm statement as to the long-range radon daughter build-up during transport of the plume. It may well be that over distances of a few kilometers and time periods of an hour or more, that the radon daughters do approach equilibrium with their radon parent which has come from the mine exhaust vents.

To determine if this is actually true would require some additional measurements. We recommend that as time and funding permit, a significant research program be initiated to determine the ratio of radon to its daughters at considerable distances from mine exhausts or stockpiles and tailings piles in the uranium mining and milling areas. This information is needed to determine in an absolute way what the local and regional effect of radon release from mine ventilation air and from other sources is.

TASK 3 - INCENTIVES FOR USING ELEVATED RELEASE POINTS TO REDUCE GROUND LEVEL RADON CONCENTRATIONS FROM UNDERGROUND URANIUM MINES

The conclusions drawn are based on a reasonably small data base. Although some general guidance relative to the effects of elevated releases is offered, more definitive information could be generated on a site-specific basis, particularly in the case of strongly biased distributions of wind direction and/or population. Conclusions drawn with the aid of the Hanford and Black Fox data bases are:

- Despite differences in meteorology, different sites can have very similar annual average distributions of concentration.

- Despite similarity of magnitude and distribution of ground-level concentration resulting from ground-level releases, the two sites displayed quite different ground-level concentrations following modeled elevated releases. A greater reduction in concentration was associated with the site with more stable atmosphere (Black Fox).
- A 20-m release elevation reduces annual average concentration (when compared to a ground-level release) by about 60% at one mile from a source, and by about 30% at ten miles from the source.
- The axis of maximum benefit from elevated releases cannot be specified on the basis of the concentration distribution generated by a ground-level release.
- The height of the mixing layer has a relatively small influence on the benefit accruing as the result of elevated releases. Specifically, the elevated release benefit is about the same for a modeled 200-m inversion "lid" or a 1000-m inversion "lid."
- Benefits, in terms of reduced dosage to population per unit of stack height increase, decrease as stack height increases. This conclusion is documented in Figure 9.

  Beyond roughly 1.5 miles from the centroid of up to three 20-m stacks (spaced a mile or less apart), the effect of stack locations is minimal.

Recommendations

The scope and funding of this task did not permit an examination of concentrations resulting from a large number of the possible combinations of site meteorology, source configurations, source heights, and atmospheric mixing heights. If one were to evaluate this technique in principle for all underground uranium mining sites, a large variety of site and meteorology parameters would need to be considered. By studying a major number of underground mines in this manner, one would be able to intercept the impact of using elevated releases to reduce ground-level radon concentrations from the entire underground uranium mining industry. The data generated from this study indicate that the effects of elevated releases are very site-specific. This type of study is particularly applicable to assess the radon dose received by receptors in the vicinity of a mine.

TASK 4 - A STUDY OF THE CONTRIBUTION OF MULTIPLE RADON SOURCES IN THE AMBROSIA LAKE DISTRICT, NEW MEXICO, TO CONCENTRATIONS AT SELECTED LOCATIONS

Uranium mining and milling operations result in a multitude of radon releases of varying source strength. An analysis is performed of the relative importance of these various radon releases on ambient atmospheric radon values. A uranium mining and milling region is selected to provide case study data on the locations, numbers, and magnitudes of releases. A comprehensive list of radon releases is compiled. Then using the best available information, source terms are generated for each location. Estimates of the atmospheric transport and dispersion of these emissions provide a framework for comparison of the relative contributions of atmospheric radon. The mine vent radon releases contribute a much larger fraction of the incremental atmospheric radon values than tailings piles. Although a few mine vents do have large radon emission rates, the large number of smaller mine releases constitutes a major fraction of total radon emissions.

Recommendations

The source term data have clearly pointed out the importance of the vent releases in total radon incremental concentrations. Future studies should have stronger emphasis on the vent releases. The models should account for actual configuration of vent releases (size, elevation, exit velocity, etc.). The assumption of ground-level releases may be resulting in considerable overestimation of short-range concentrations.

Verification of the modeled concentrations should be made using monitoring data and onsite studies of specific dispersion conditions.

The results should be generalized to other regions by comparisons of the vent configuration, numbers, and locations. Should the annual radon concentrations be limiting, similar efforts should be undertaken for other regions.

TASK 1

EFFECT OF BULKHEADING OR FILLING UNDERGROUND MINES ON RADON EMISSION*

INTRODUCTION

Bulkheading in underground mines is a common practice, and it is performed with the objective of controlling ventilation air to maximize air quality to which miners are exposed, while minimizing the total amount of air movement required. In a very practical sense, bulkheading is essential for many underground mining operations to permit the proper ventilation of working areas. In some cases, bulkheading may effectively exclude major sections of underground uranium mines from the ventilation pattern. There are, however, problems involved in bulkheading, the major one being that there can be leakage from the bulkheaded area back into the working area. The radon concentrations behind bulkheads may be orders of magnitude higher than those in work areas, and such a leakage could have serious consequences on exposure to mine workers. For this reason, it is common practice to maintain a bulkheaded area at a slightly negative pressure. The exhaust ventilation rate required to maintain the slight negative pressure behind the bulkheads is small compared to the additional ventilation which would be required without the bulkheading. This assures that any leakage through the bulkheads will be from the working areas to the bulkheaded areas and should, therefore, prevent unexpected high exposures in working areas. When the bulkheaded area is kept at a slightly negative pressure (nominally 0.03 cm water) with respect to the active areas, the air being pumped from behind the bulkhead which contains very high radon concentrations can be released to the environment outside the mine or passed through a radon sorption trap prior to release.

While the bulkheading operation provides an important function in permitting practical ventilation of underground working areas, it also probably

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*This work was carried out jointly with the U. S. Bureau of Mines. An open literature publication entitled "Bulkheading Effects on Radon Release from the Twilight Uranium Mine" has been prepared by V. W. Thomas, PNL, Richland, WA, and C. S. Musulin and J. C. Franklin, U. S. Bureau of Mines, Spokane Research Center.

has a significant effect on the total radon released to the atmosphere. The objective of this current study is to determine what the magnitude of this effect is during both exhausting and blowing ventilation.

EXPERIMENTAL PROTOCOL

The Twilight Mine (Figure 1) just north of Uravan, Colorado, was selected as the site for the investigation. This mine is leased from Union Carbide by the Bureau of Mines and is operated as a research mine for the Bureau of Mines by the Adare Contracting Company. The Twilight Mine is small, with two entry portals connected to each other by a single loop drift of about 360 m in length (Figure 1). Two internal loops have been mined off from this main loop. The larger of these, which is about 250 m in length, is equipped with bulkheads at either end. Each end can be opened or closed by doors. The bulkhead at 1L9 is penetrated by two small diameter pipes (5 cm and 8 cm) which are normally used to pump water and/or diesel fumes into the bulkheaded portion of the mine for experimentation purposes. In this study one of these pipes was used to provide a negative pressure of about 0.03 cm of water behind the closed bulkheads with respect to the rest of the mine. A larger (60 cm) pipe penetrates both bulkheads and can be opened or closed by means of louvered shutters.

The main ventilation fans are located at Portal B. They may be reversed to provide either exhausting ventilation or blowing ventilation. In the experiment, exhausting ventilation rates averaged 390 m^3/min and blowing ventilation averaged 550 m^3/min. The experiment was conducted in four major phases using two modes of ventilation:

Phase I:	Exhausting ventilation with bulkhead doors open
Phase IIA:	Exhausting ventilation with bulkhead doors closed, 60 cm pipe shutters open (leaky bulkhead)
Phase IIB:	Exhausting ventilation with bulkhead doors closed, 60 cm pipe shutters closed
Phase IIIA:	Blowing ventilation with bulkhead doors closed, 60 cm pipe shutters open (leaky bulkhead)
Phase IIIB:	Blowing ventilation with bulkhead doors closed, 60 cm pipe shutters closed

Phase IV: Blowing ventilation with bulkhead doors open

Measurements were made during the course of this investigation using instruments provided and operated by the Bureau of Mines, Spokane Research Center (SRC), in addition to those installed by Pacific Northwest Laboratory (PNL). The monitoring equipment was operated and maintained during the course of the study by the Adare Contracting Company.

The following continuous measurements were made throughout the four phases of the experiment in addition to those at the exhaust portals (see Figure 1).

Station 1 - Radon concentration, air velocity
Station 2 - Radon concentration, air temperature, barometric pressure
Station 3 - Radon concentration
Station 4 - Radon concentration
Station 5 - Radon concentration, air velocity
Station 6 - Radon concentration

This monitoring instrumentation located at the above stations is described elsewhere by Franklin.[1] In addition, two Eberline Continuous Radon Gas Monitors, Model RGM-1, were used to measure radon concentrations at Stations 0 and 7 and at bulkheads 1L7 and 1L9, dependent upon the experimental conditions. Vane anemometers and hot-wire anemometers were used to make spot measurements of air velocities at selected places in the mine.

This mine is situated about 30 m below the top surface of a mesa. Measurements of radon flux from the ground above the bulkheaded area were made during 24-hour periods with and without the bulkheads being closed to check for increased radon flux at the surface due to possible faults or fissures from the mine. A fissure reaching to the surface from the bulkheaded area would be expected to emanate much more radon than the surrounding areas, especially when the bulkhead doors were closed and without negative pressure in the bulkheaded area.

Radon flux measurements from the surface of the mesa above the mine were made using a charcoal canister technique similar to that reported by Countess[2] (GMA 457315 canister: Mine Safety Appliance Co.). An array of

canisters was set out in a grid pattern starting at drill hole J316 and the canisters were separated by about 30 m (Figure 2). The canisters were sealed to the ground by pushing a cylindrical edge into the ground 1 to 2 cm. Exposure times were 24 hours. After exposure, the canisters were sealed and shipped to our laboratory for ^{222}Rn measurement based on gamma-ray spectrometric analysis for its ^{214}Bi and ^{214}Pb daughters.

In addition to measuring radon concentrations, 400 g quantities of an inert tracer (freon-12) were released into mine inlet air during each of the four phases of ventilation described earlier to measure the time required to transport gases from portal to portal and their streaming effects. For these analyses, samples of the mine exhaust air were collected at timed intervals after each release and analyzed for freon-12 concentrations by gas chromatographic techniques. Measurement of freon in the air being pumped from behind the bulkhead served to indicate the degree of leakage across the bulkhead.

DISCUSSION AND CONCLUSIONS

The freon release experiments described above provided two useful pieces of information. First, it was learned that a very slight negative pressure (0.03 cm water) in the bulkheaded region caused mine air to leak from the "active" area of the mine into the bulkheaded area. Relative concentrations of freon-12 measured in air being pumped from behind the bulkheaded area during portions of Phase IIIA and Phase IIB are presented in Figure 3. The freon released in the active portion of the mine was, within minutes, detected in the air being pumped from the bulkheaded area. Second, after a single spike release at the intake portal under each of the four sets of ventilation conditions described earlier, the time required to reduce the concentration of freon-12 at the outlet portal by 50% was half as long with the bulkhead doors closed as with them open (Figures 4 and 5). Removal of 95% of the freon tracer took three to four times longer with the doors open than when closed. The data indicated, at least for 50% removal of the tracer, that clearance time under exhausting ventilation was faster than under blowing ventilation.

Radon flux measurements on the surface of the mesa above the bulkheaded portion of the mine were comparable during a 24-hour period when the bulkhead

doors were open under exhausting ventilation, and a 24-hour period when the bulkhead doors were closed under blowing ventilation. The small changes in radon flux which were observed are probably attributable to normal variations between days. The flux measurements obtained are presented in Table I. No large changes in flux of the magnitude to be expected from possible fissures were observed though average radon concentration behind the bulkhead doors was a factor of 300 higher when the bulkhead doors were closed than when they were open. Thus the integrity of the mine appears sound.

Figures 6 through 11 compare barometric pressure and the quantity of radon leaving the exit portals during the six study periods. Diurnal variations in radon concentrations are easily recognized as are inverse correlations of radon concentration with barometric pressure changes. There are also some discontinuities in the data due to inadvertent ventilation fan shutdowns and during stabilization periods after changing experimental conditions from one phase to another. The data are sufficient, however, to clearly show the effects of the bulkheading procedure.

During exhausting ventilation with the bulkhead doors open (Phase I), the radon leaving the mine averaged 75 μCi/min (Table II). Closing the bulkhead doors but leaving the 60 cm pipe shutters open (leaky bulkhead) reduced that average amount by 17% to 62 μCi/min. Closing the 60 cm pipe shutters (Phase II) resulted in a further reduction of 35% for a total of 52% reduction from the open bulkhead condition. As mentioned earlier, the radon concentration behind the bulkhead had not come to an equilibrium state by the time Phase IIB measurements were made. At equilibrium, the radon concentration behind the bulkhead might be 5 to 10 times higher which would certainly change the 52% reduction in radon to a smaller figure, possibly on the order of 30% to 40%. In addition, while the average barometric variations during Phases I and IIA are comparable, the barometric pressure went through a very steep increase just prior to Phase IIB and remained at a relatively high pressure throughout Phase IIB. This may tend to reduce radon emanation from the mine and thus reduce radon leaving the mine. It was found that closing the bulkhead doors and the shutters did not completely isolate the bulkhead area, and so these conclusions describe a bulkhead with small leaks. Even with this situation, there is a real and important reduction in radon release to the environs.

TABLE 1. Radon Flux Measured on Mesa Above Twilight Mine, Uravan, Colorado

SAMPLE	^{222}Rn, $pCi/cm^2/sec \pm 1\sigma$ *	
	EXHAUSTING VENTILATION, DOORS OPEN	BLOWING VENTILATION, DOORS CLOSED
NW-2	7.4×10^{-5}	1.2×10^{-4}
NE-2	$9.8 \times 10^{-6} \pm 4.6 \times 10^{-6}$	$4.4 \times 10^{-5} \pm 5.3 \times 10^{-6}$
NW-1	2.2×10^{-5}	$4.0 \times 10^{-5} \pm 6.0 \times 10^{-6}$
N-1	2.4×10^{-5}	$4.3 \times 10^{-5} \pm 5.7 \times 10^{-6}$
NE-1	6.3×10^{-5}	$1.9 \times 10^{-5} \pm 4.8 \times 10^{-6}$
NW-0	7.5×10^{-5}	4.1×10^{-5}
N-0	9.3×10^{-5}	1.4×10^{-4}
N-V**	3.8×10^{-3}	Not sampled
NE-0	$5.2 \times 10^{-5} \pm 5.4 \times 10^{-6}$	$4.6 \times 10^{-5} \pm 6.3 \times 10^{-6}$
SW-0	9.0×10^{-5}	$7.4 \times 10^{-5} \pm 6.7 \times 10^{-6}$
S-0	4.3×10^{-5} 4.9×10^{-6}	$6.1 \times 10^{-5} \pm 6.0 \times 10^{-6}$
SE-0	7.0×10^{-5}	1.4×10^{-4}
SW-1	6.4×10^{-5}	1.8×10^{-4}

* Counting uncertainties less than 5% unless given

** Old drilling hole (J-316) adjacent to N-0

Table 2. AVERAGE RADON IN VENTILATION AIR FROM THE TWILIGHT MINE

Experimental Conditions	Measurement Period (days)	Exhaust Air from Bulkhead Area		Exhaust Air from Total Mine		Ratio of Bulkhead Area Radon to Total Mine Radon
		Flow Rate m^3/min	^{222}Rn µCi/min	Flow Rate m^3/min	^{222}Rn µCi/min	
Phase I Exhausting Ventilation Bulkhead Doors Open	16	66	42	390	75	0.56
Phase IIA - Exhausting Ventilation Leaking Bulkhead	11	None	--	390	62	--
Phase IIB Exhausting Ventilation Bulkhead Doors Closed	0.15	1.5	2.5	390	36	0.07
Phase IIIA Blowing Ventilation Leaking Bulkhead	6	None	--	560	84	--
Phase IIIB Blowing Ventilation Bulkhead Doors Closed	5	1.5	1.2	540	55	0.02
Phase IV Blowing Ventilation Bulkhead Doors Open	13	58	30	560	79	0.38

During blowing ventilation with the bulkhead doors open (Phase IV), the radon leaving the mine averaged 79 μCi/min. Closing the bulkhead doors but leaving the 60 cm pipe shutters open (leaky bulkhead) (Phase IIIA) resulted in an apparent increase of 6% from the open-door condition to 84 μCi/min. We do not consider this change to be significant as it may be due to average barometric change differences between the two periods. Closing the 60 cm pipe shutters (Phase IIIB) reduced the average total radon leaving the mine to 55 μCi/min for a total reduction of 30% from the open-door condition.

When the bulkhead doors were open, exhausting ventilation resulted in an average radon emission of 5% less than blowing ventilation. We feel that this small difference is not significant and may simply be due to minor differences in the average barometric pressure.

To understand the significance of the radon measurements in Table II, one must again consider the lengths of the experiment during which radon was measured. All of these time periods except one ranged from 5 to 16 days, and thus the ratio of radon being released from the bulkheaded area relative to the total mine was probably representative. In Phase IIB, however, the period of this experiment was only about 3-1/2 hours. During this period of time, the bulkhead doors and the 60 cm pipe shutters were closed but the radon in the bulkheaded area had not begun to approach equilibrium. If the experiment had been allowed to continue for several days, the amount of radon being exhausted from the bulkheaded area would certainly have increased. To place this in perspective, we have estimated the approximate amount of air in the bulkheaded area. We estimate this amount may be on the order of 5000 m^3. Since the ventilation rate from the bulkheaded area is 1.5 m^3/min, then this would be equivalent to a daily exchange of the air. One could, therefore, expect a considerably larger build-up in radon concentration than was observed during the 3-1/2 hour period of the test.

In Phases IIA, IIIA, and IIIB of the experiment, measurements were conducted over periods of from 5 to 11 days and, therefore, the radon release from the bulkheaded area would more closely represent that at equilibrium.

Phase IV was simply a test with the bulkhead doors open, and the data for this were accumulated over a 13-day period and should certainly be

representative. The important thing to recognize from these experiments, however, is that the radon which is discharged from behind the bulkheaded area at relatively low flow rates could be discharged by use of sorption traps if the economics were favorable* or it could be released at a relatively high altitude, say 15 to 25 m. In either case, the opportunity for exposure to individuals at ground level would be reduced.

As can be seen from the data in Table II with bulkhead doors closed and with exhausting IIB and blowing IIIB ventilation, a very major reduction in radon release would be achieved if the radon from the bulkheaded region were captured on a sorption trap. For these experimental conditions, the total radon released from bulkheading and trappping would be 45% and 68% of the nonbulkheading situation for exhausting and blowing ventilation, respectively.

SUMMARY

A study of the effects of bulkheading on the radon release from an underground uranium mine was conducted at the Bureau of Mines underground uranium research mine. The research mine is known as the Twilight Mine and is located near Uravan, Colorado. The air flow conditions included exhausting and blowing ventilations with and without bulkheading for six consecutive experimental periods of up to two weeks each. Radon measurements in mine and exhaust air were made for both directions of air flow from bulkhead doors at either end of an internal loop open, closed, and closed but with a major leak. These measurements show that bulkheading produced a major effect on the total radon leaving the mine and that this effect was dependent upon the type of ventilation and the integrity of the bulkheads. During exhausting ventilation, the total radon discharged from the mine decreased by 17% when the bulkhead doors were closed but with a large leak. A further reduction of 35% was observed when the large leak was closed. This latter measurement, however, was made over a very short time period and

*The technical aspects of the sorption of radon on charcoal beds appear to be favorable[3], but it is beyond the scope of this study to evaluate the economical feasibility of the procedure.

radon had not built up to its maximum equilibrium concentration. During blowing ventilation, a small (6%) but not significant increase in the total radon leaving the mine was measured when the bulkhead doors were closed but with the large leak. When the large leak was eliminated, a reduction of 35% was observed. Other parameters affecting the total radon leaving the mine, including barometric pressure, must also be taken into consideration when interpreting these measurements. A small amount of air was pumped from the bulkheaded area of the mine to maintain a negative pressure. This air, with its relatively high radon content, could be passed through a sorption trap or discharged at a high elevation to minimize exposure to people in the environs. If this air were passed through absolute sorption traps, it could reduce the radon leaving the mine to 45% and 68%, respectively, of that for exhausting and blowing ventilation with no bulkheading.

RECOMMENDATIONS

While valuable information was obtained from this bulkheading experiment, it is recognized that a large-scale experiment should be conducted in an operating uranium mine. Battelle scientists have met with the Bureau of Mines and with representatives of a major uranium mining company to discuss both the current experiment and a possible experiment in the future at a large uranium mine.

It seems important to compare the potential of bulkheading in a large active mine with the observations which have been made in this very small-scale study. Whereas the experimental mine had somewhat comparable areas of active and inactive regions, an actual working mine may have worked-out regions with tens of times more volume than the working regions. It can thus be surmised that effective bulkheading of active mines could reduce radon emissions by up to an order of magnitude, or perhaps more. If a study were to be conducted at an active uranium mine, one should be selected where the potential for bulkheading of a substantial region of the mine volume is possible. Discussions with mine operators have indicated their willingness to participate in a study if a well-conceived experiment could be devised. From considerations of active mines, it appears that suitable ones are available and that a study which would include the cooperation of the mine operator, the Bureau of Mines, and PNL could be productively carried out.

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Fusamura, N., Kurosawa, R., and Ono, S., "On the Study of Radon Removal with Active Carbon," Nippon Nogyo Kaishi 79:590, August 1963.

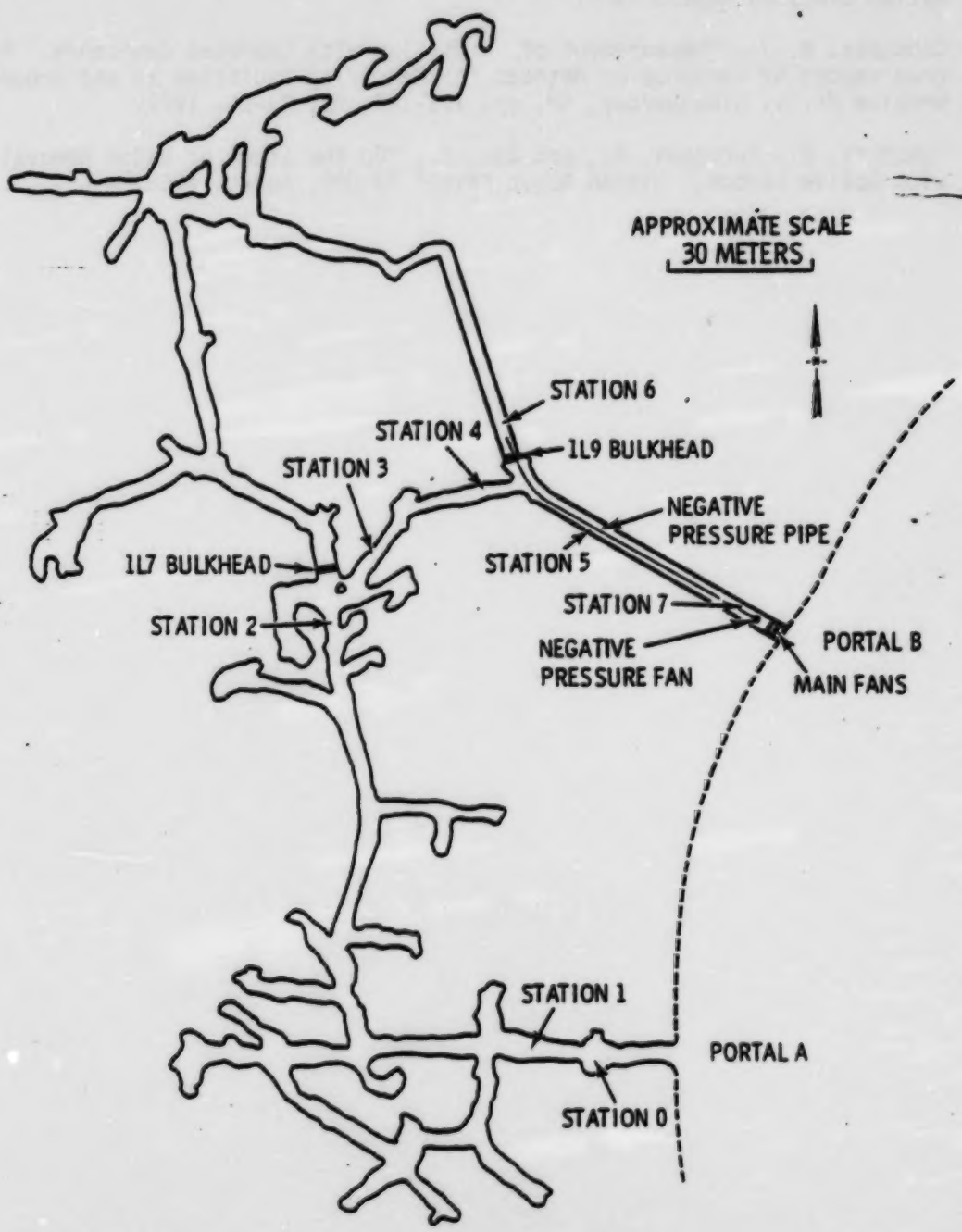

Figure 1. Twilight Mine, Uravan, Colorado.

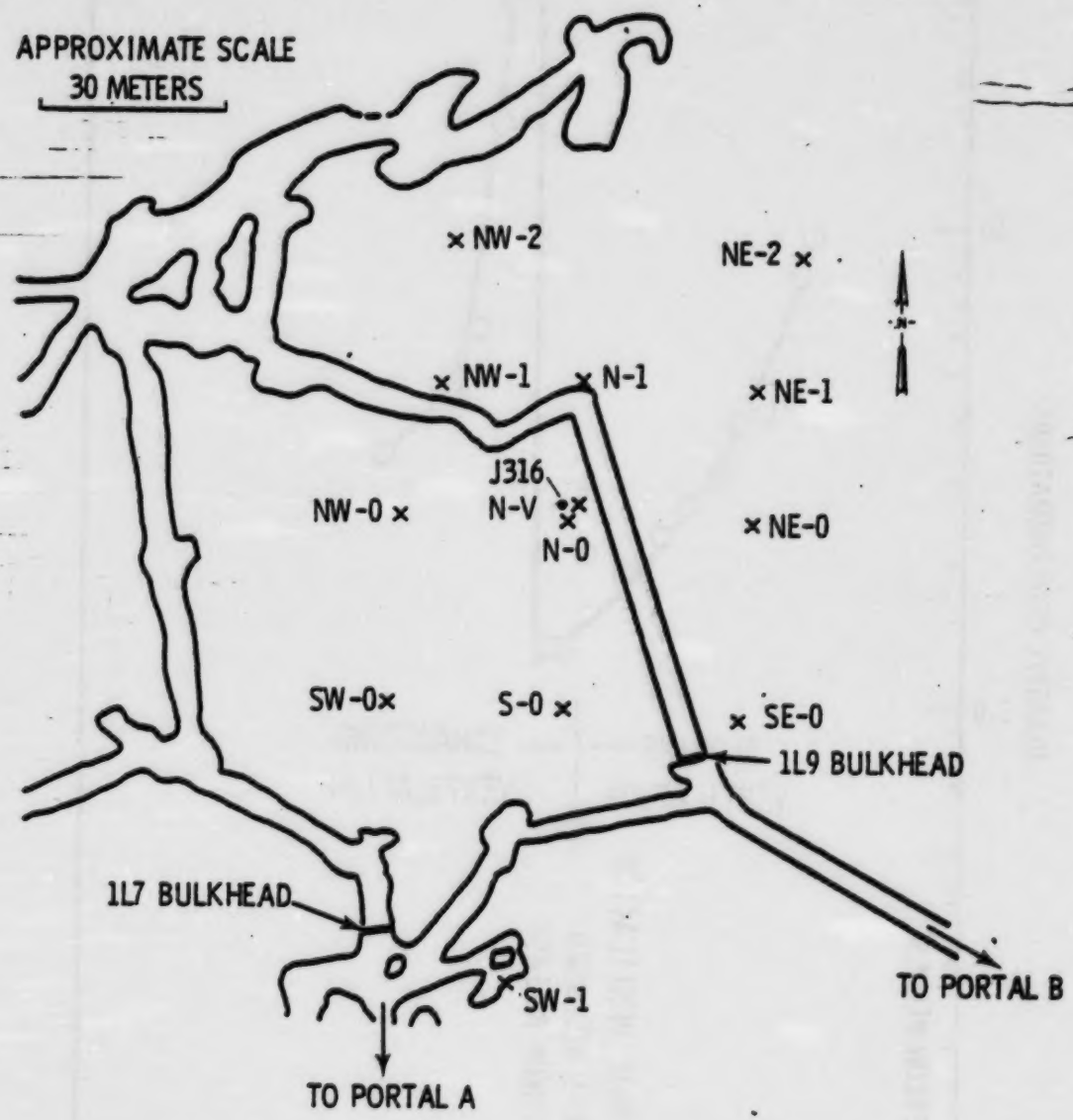

Figure 2. Approximate Radon Flux Sampling Sites on Mesa Above Twilight Mine

Depth from Mesa to Mine = 30 Meters

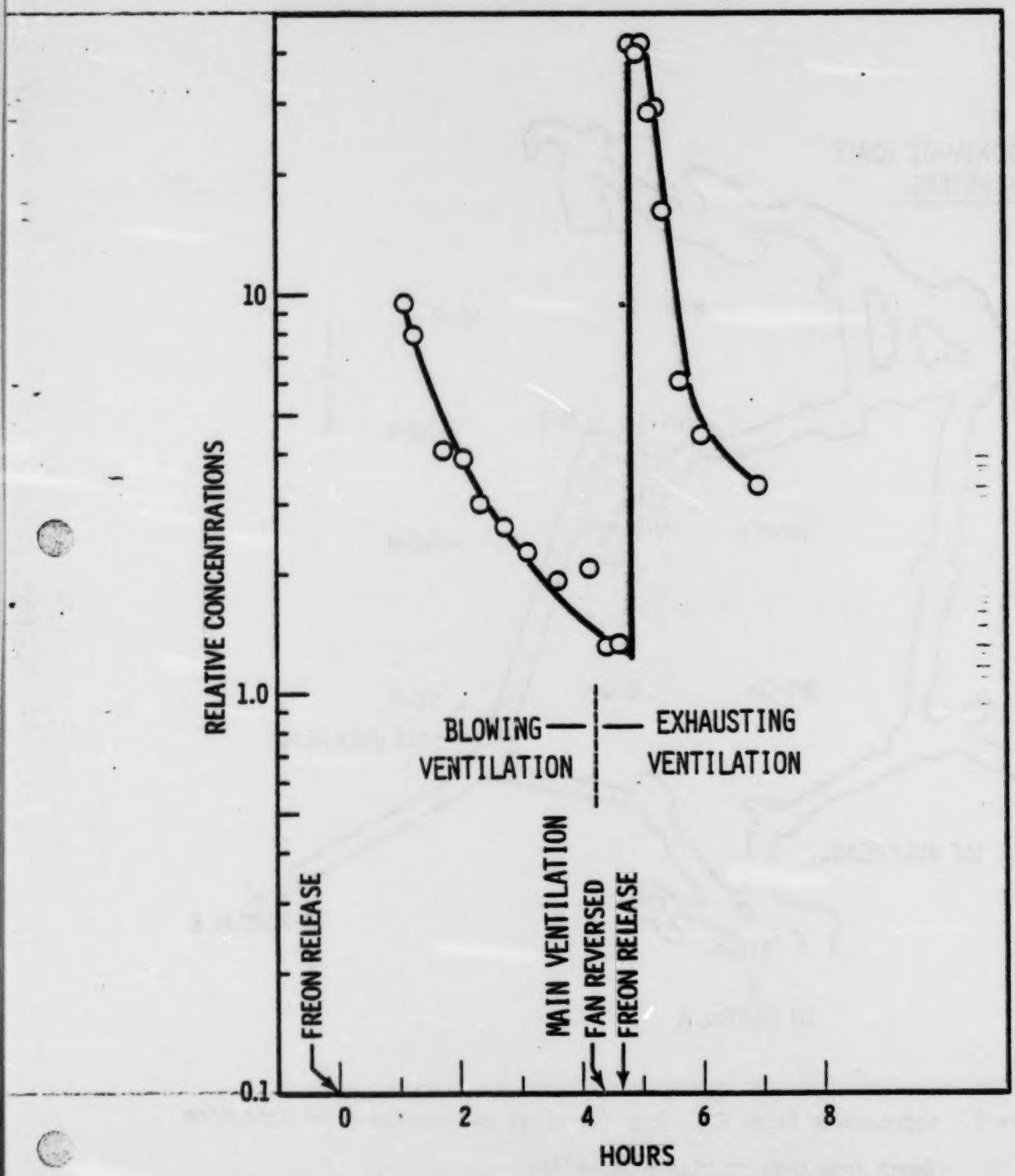

Figure 3. Freon Concentrations in Air Being Pumped from Behind the Bulkheaded Area During Portions of Phase IIIA and Phase IIB

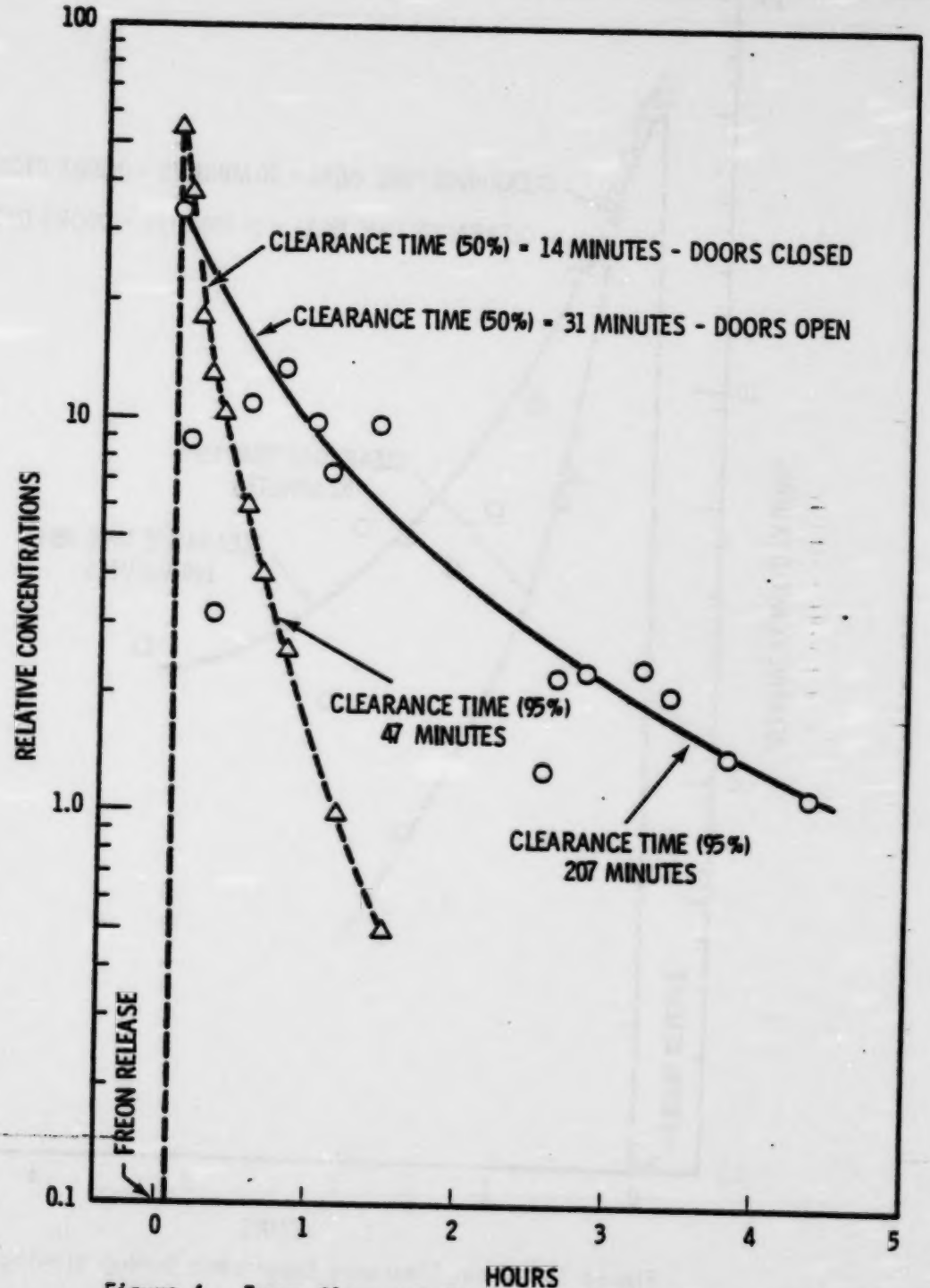

Figure 4. Freon Clearance Experiment During Exhausting Ventilation.

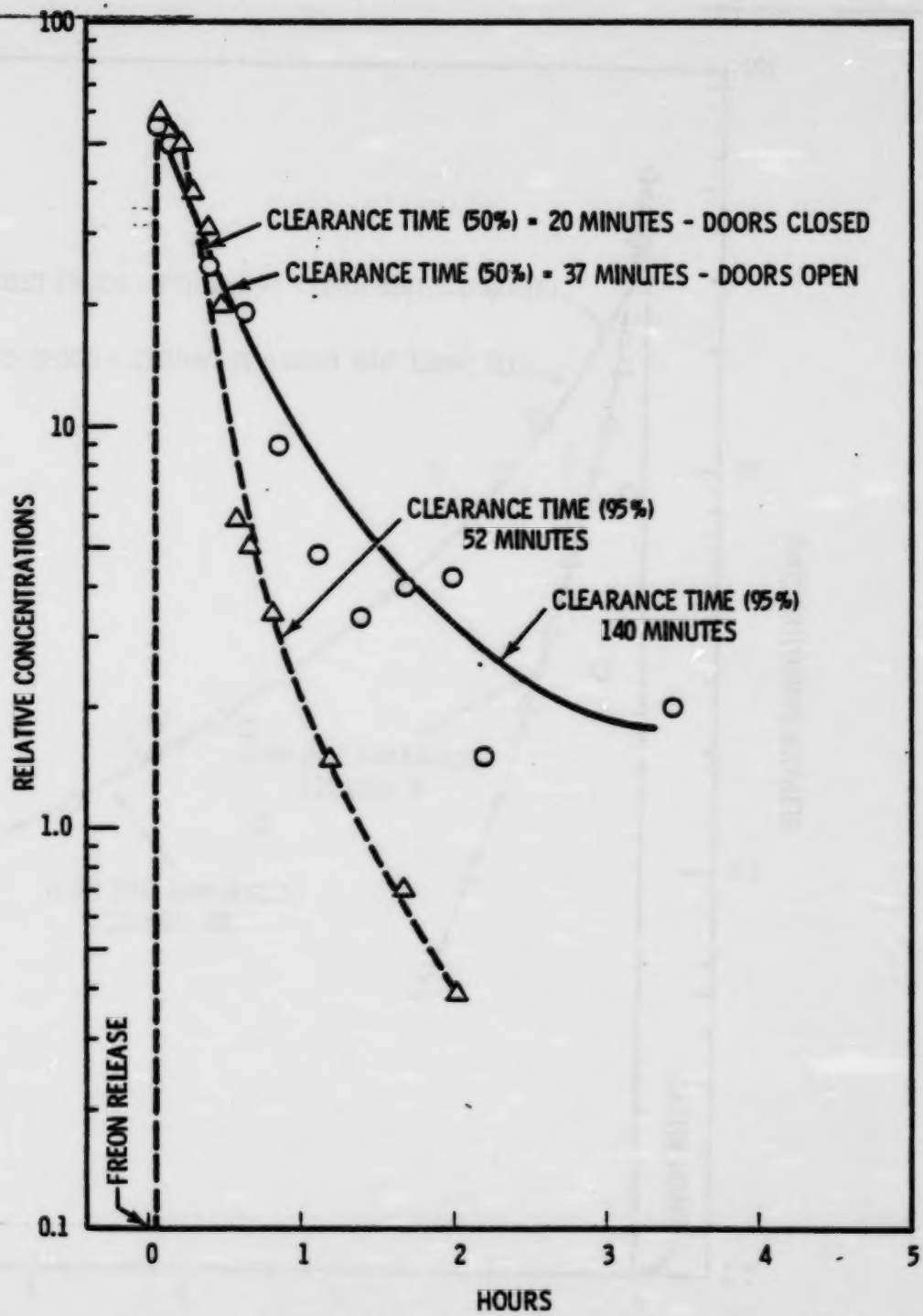

Figure 5. Freon Clearance Experiment During Blowing Ventilation.

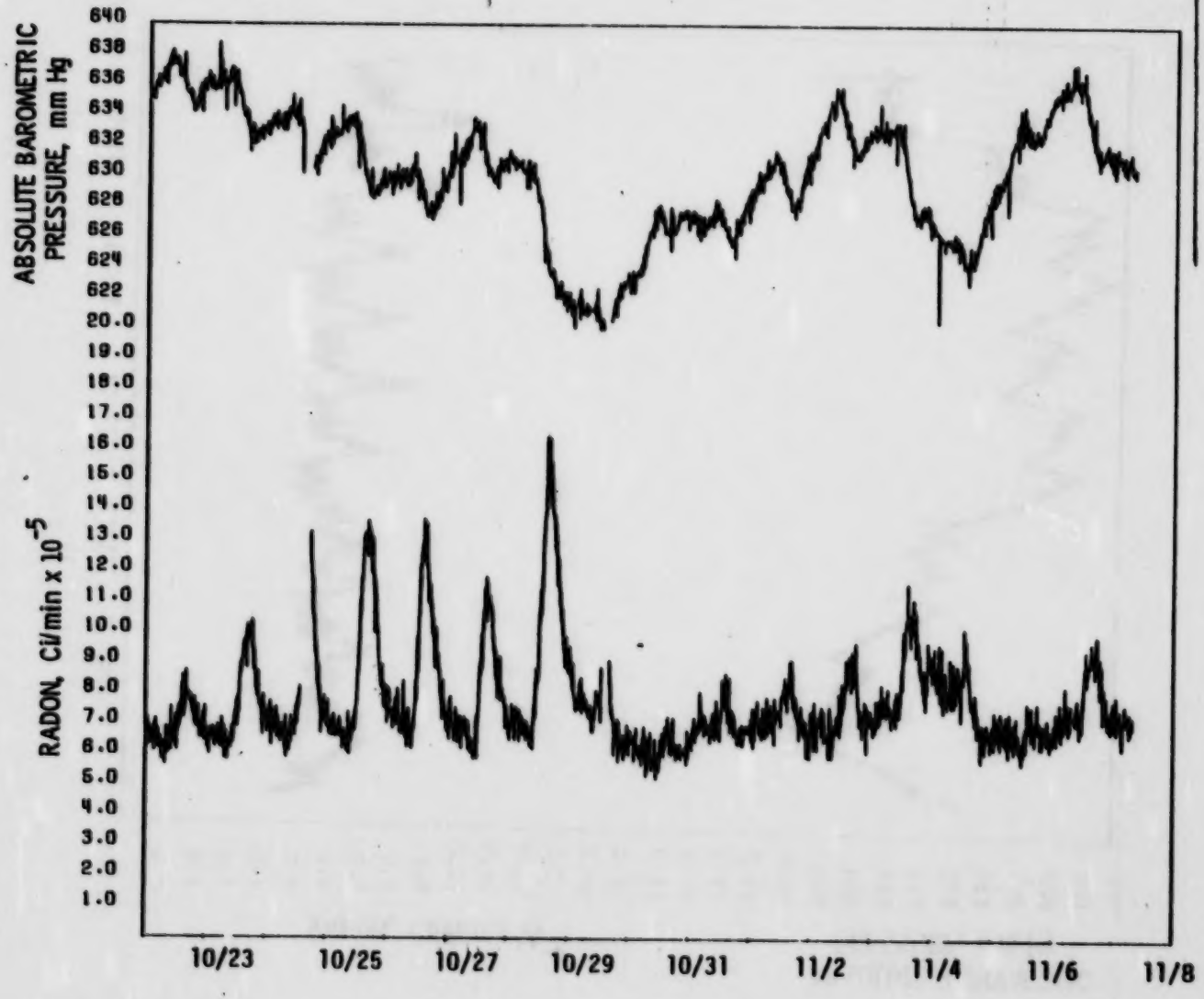

Figure 6. Phase I, Exhausting Ventilation, Doors Open

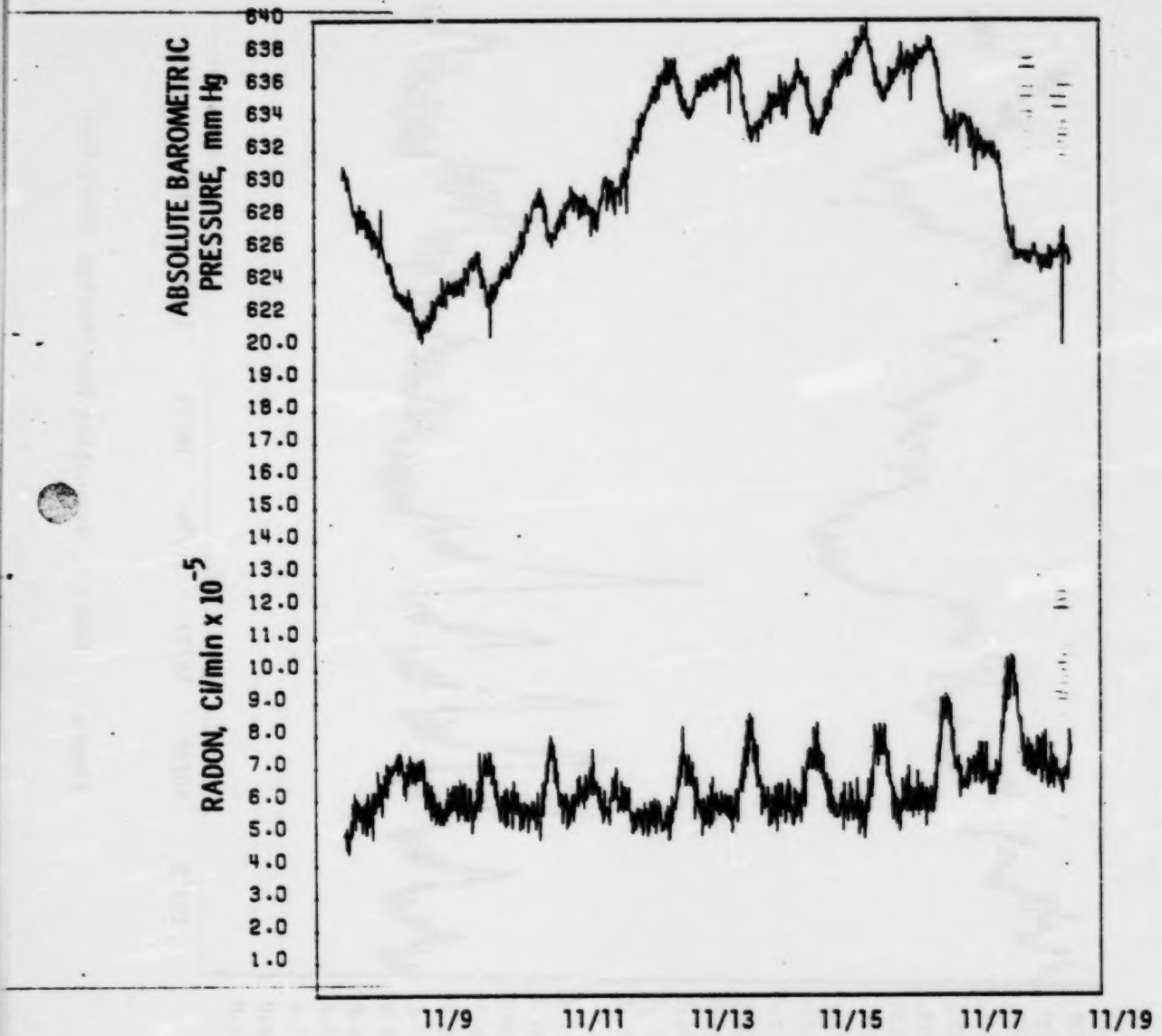

Figure 7. Phase IIA, Exhausting Ventilation, Doors Closed, 60 cm Pipe Shutters Open.

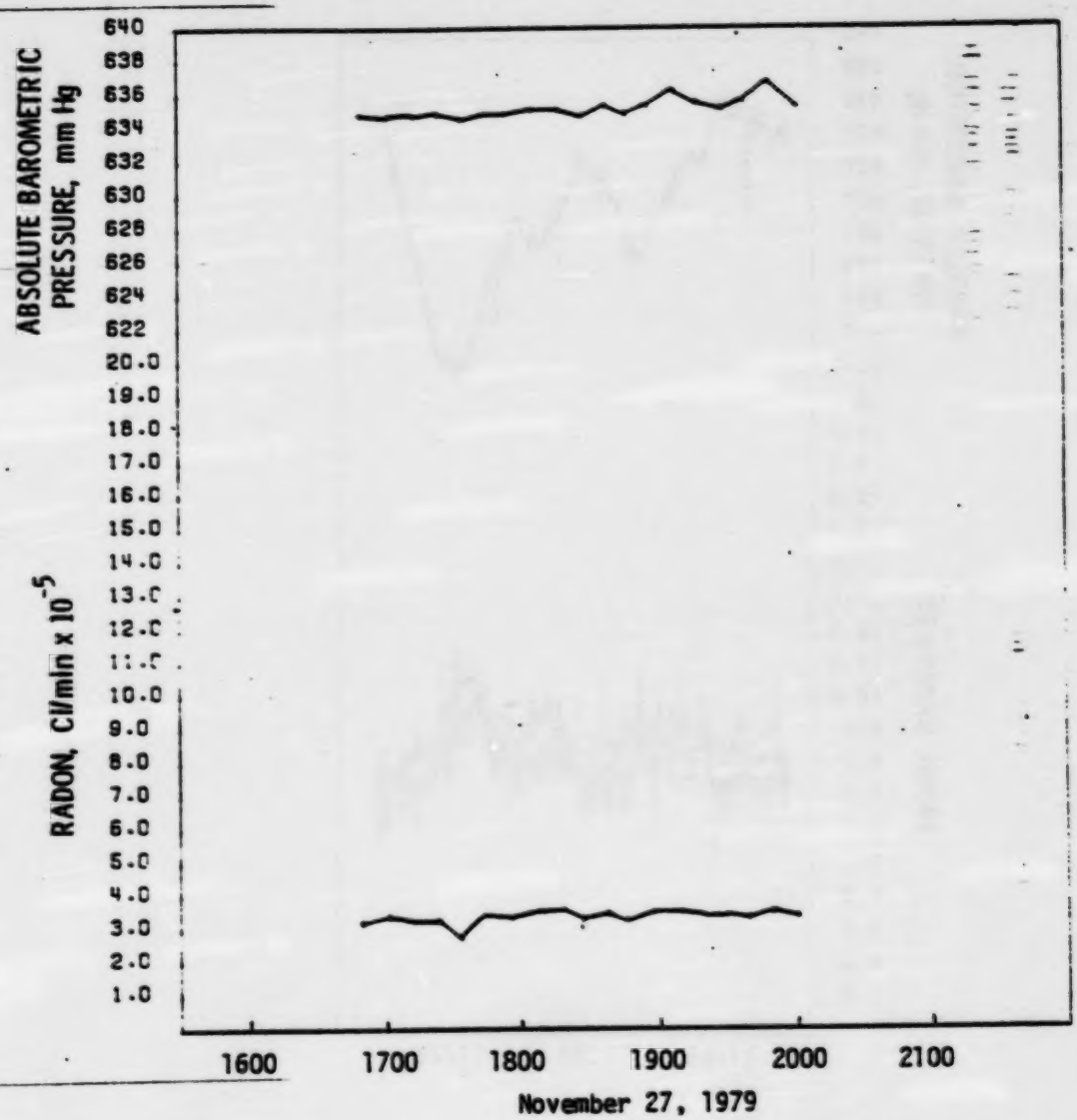

Figure 8. Phase IIB, Exhausting Ventilation, Doors Closed, 60 cm Pipe Shutters Closed.

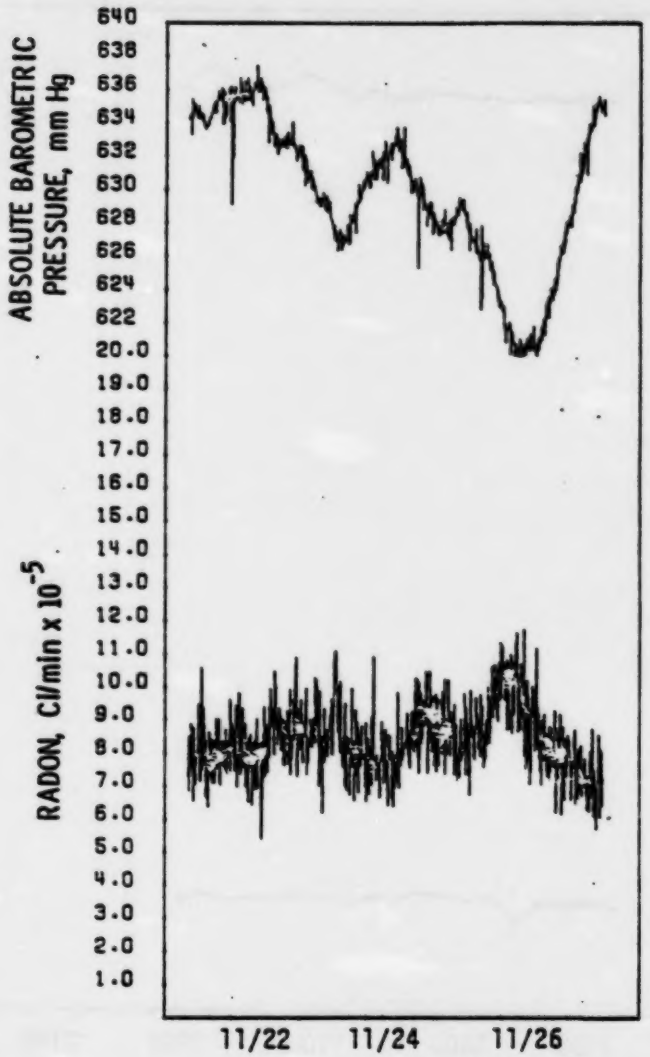

Figure 9. Phase IIIA, Blowing Ventilation, Doors Closed, 60 cm Pipe Shutters Open.

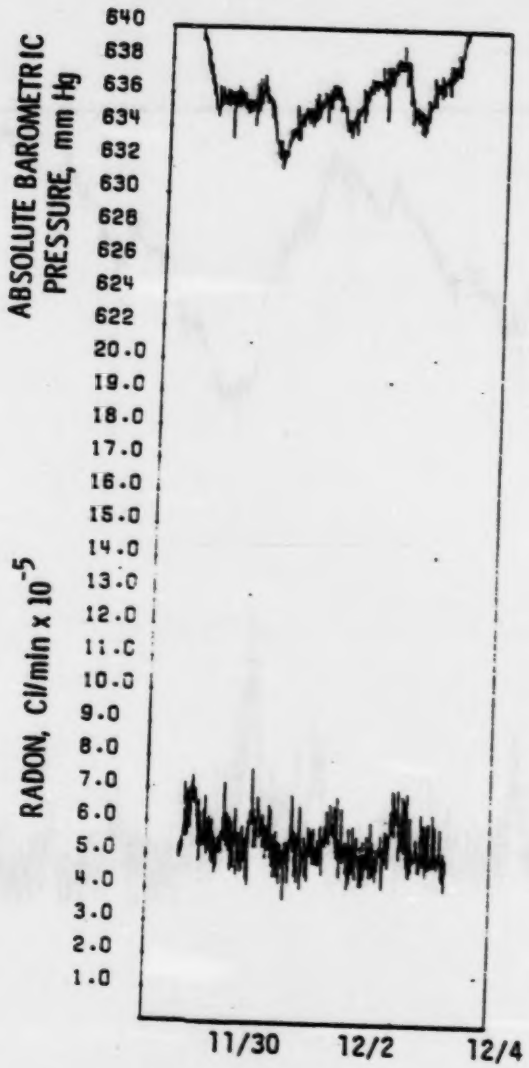

Figure 10. Phase IIIB, Blowing Ventilation, Doors Closed, 60 cm Pipe Shutters Closed.

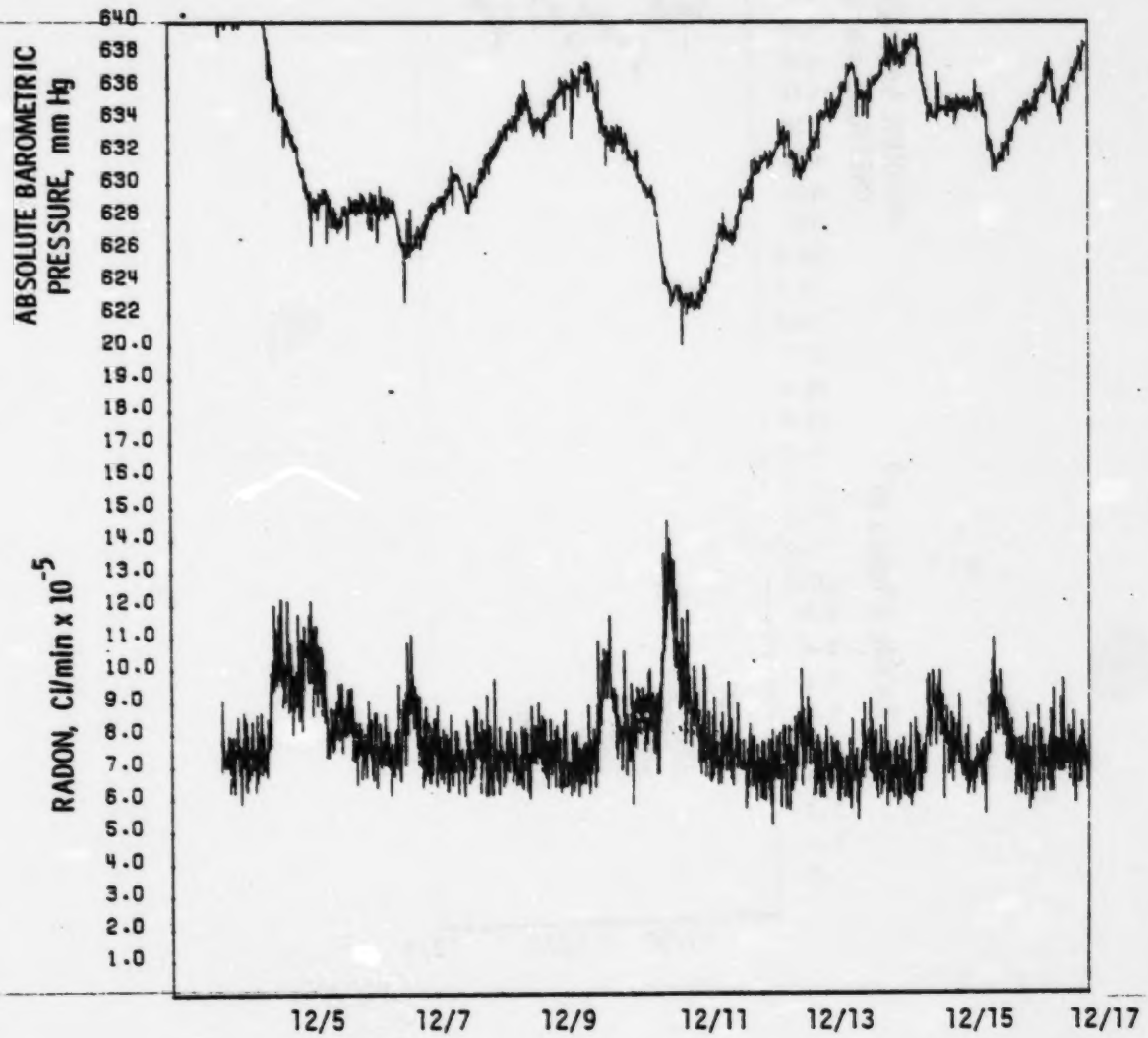

Figure 11. Phase IV, Blowing Ventilation, Doors Open.

TASK 2

RADON DAUGHTER EQUILIBRIUM AT VARIOUS DISTANCES FROM EXHAUST VENTS OF UNDERGROUND URANIUM MINES

INTRODUCTION

The exposure to individuals in the vicinity of a uranium mine vent depends to a large degree on the ratio of radon daughters to radon. Exposures to mine workers are expressed in terms of working levels where one working level is defined as any combination of radon daughters which releases 1.3×10^3 MeV of energy per liter of air. It is also defined as that amount of energy which would be released by the decay of radon daughters which were in equilibrium with 100 pCi/ℓ of radon.

EXPERIMENTAL PROTOCOL

In this experiment, the concentrations of radon and radon daughters were measured at two vents of a major mine operation in the Ambrosia Lake, New Mexico area. One of these was a vertical vent with air being exhausted straight into the air. The second was a horizontal vent with the exhaust being ejected parallel to the ground. In our discussions with the mining engineers, it appears that the horizontal vent will be more typical of future operations since this type of vent eliminates the heavy fan bearing wear which results where vertical exhaust installations are used. These vents were chosen because they were upwind from local tailings piles and because they had previously been found to emit higher concentrations of radon than other vents in this area. These factors serve to maximize the signal from the source relative to ambient background levels.

Some of the vent exhausts were extremely wet, discharging tens of liters of water and mist per minute with the air. This resulted from the leakage of underground water through the exhaust vent casing from aquifers above the base of the mine exhaust channel. Current plans call for the sealing of vent casings to exclude water from the air stream.

Ambient wind directions were checked using a puff of smoke. All sampling was conducted in the apparent downstream plume of the vent. However, it was not uncommon for the wind to shift slightly during a run. Samples were collected sequentially at the vent and at three distances downwind.

DISCUSSION

In our experiments, we passed 140 liters of air during a 5-minute period through a fiberglass filter paper which was backed by dual charcoal traps. The final trap was held at dry ice temperature. The radon daughters which are attached to particulates were collected on the air filter, and the radon was quantitatively collected on the charcoal traps. The air filters were analyzed immediately to determine RaA, RaB, and RaC by taking consecutive 2-minute, 8-minute, and 8-minute counts on a portable alpha particle counting system.[1] Radon daughters are computed from the three alpha counts using an integral form of the Bateman equations. The radon which was collected on the charcoal trap was measured a few days later at our laboratory using our multidimensional gamma-ray spectrometer. The uranium dust contribution to the observed count rates was shown to be negligible in measurements reported in "An Environmental Study of Active and Inactive Uranium Mines, Mills, and Their Effluents, Part 2," Task 2. The radon and radon daughter concentrations observed in these experiments are tabulated in Tables 1 and 2, together with the ratios of radon to its daughters and the calculated working level exposures at the sampling sites. As indicated, measurements were made at wind speeds ranging from 1.5 to 5 m/sec for the dry vent and 1.3 to 2.7 m/sec for the wet vent. The wind speeds were not constant during any of the measurements.

Dry ventilation exhaust air from the vertical discharge vent contained 3400 pCi of radon per liter as shown in Table 1. Radon concentrations determined at distances of 30 and 140 meters downwind from the vent were not statistically different from the average ambient levels of 0.25 pCi/ℓ at this locale.[2] The elevated radon concentration detected 85 meters from the vent suggests that the plume from the vent intercepted the ground at this distance. The absence of elevated radon values at 140 meters could reflect either additional plume dilution or deflection of the plume from the sampling point by variable winds existent during the sampling period. The RaA to radon ratio at the exhaust vent was 0.32. Radium B and RaC to radon ratios were 0.21.

Because of the low radon daughter concentrations encountered at most sampling sites, it was not possible to define precisely the changes in the

ratios of each to radon from sample to sample. Since the ratios for all daughters in each instance were quite similar to those in the vent exhaust, we suspected that there was, in fact, very little change with distance as would be expected for the short transit times involved. The working levels were calculated to be 7.43 at the exhaust fan, 0.0006 and 0.0029 at sampling points 30 meters from the vent, 0.0189 85 meters from the vent, and 0.0015 140 meters from the vent. Except for the 85-meter sampling site which we believe to be representative of a plume/earth contact point, the working levels away from the vent are below the proposed EPA* standard of 0.015 working level which exists for the average annual indoor radon working level, including background.

In a second series of measurements, the concentrations of radon and its daughters were measured from a wet horizontal vent and at various distances downwind from this vent. As indicated in Table 2, elevated radon concentrations were determined 32 meters downwind from the vent, which were reduced to ambient levels at monitoring points further downwind. The ratio of radon daughters to radon in the sample collected 30 meters downwind from the vent was significantly lower than that determined at the vent. We believe that most of this is due to scavenging of radon daughters by water droplets in the vent plume. Removal of radon daughters significantly lowered the working levels at distances exceeding 30 meters from the vent.

Relative radon daughter concentrations determined at the two stations farthest downwind of the wet vent were not depleted. It is possible either that sampling was conducted outside the plume or the plume had dispersed sufficiently that discharged radon and daughter activities were indistinguishable from the ambient levels in this location.

SUMMARY

Data obtained in the study indicate that the ratios of radon daughters to radon are relatively low in exhaust air from the underground uranium mine vents. Since working levels are determined by the total concentrations of radon daughters rather than radon itself, this means that the direct

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*Federal Register, Vol. 45, No. 79, April 22, 1980

exposure to the ventilation exhaust is much less than it would be if the radon daughters more nearly approached equilibrium with radon. The radon daughter equilibrium measurements made downwind from the ventilation exhaust ports showed low ratios indicating that either radon daughters are scavenged by the aerosols that are contained in the plume or that they are scavenged by contact with the earth's surface. In many cases, the data do not suggest a significant build-up in radon daughters with time during transport of the plume at ground level. The working levels were below the proposed indoor EPA standard of 0.015 at all points exceeding 100 meters from the vent.

RECOMMENDATIONS

The measurements which have been made in this study indicate that the ratios of radon daughters to radon are relatively low in exhaust air from the vents. Since working levels are determined by the total concentrations of radon daughters rather than radon itself, this means that the direct exposure to the ventilation exhaust is much less than it would be if the radon daughters more nearly approached equilibrium with radon. Measurements downwind from the ventilation exhaust also show low ratios indicating that either radon daughters are scavenged by the aerosols that are contained in the plume, as is apparently the case for the wet exhaust, or that they are scavenged by contact with the earth's surface. In many cases, the data do not suggest a significant build-up in radon daughters with time during transport of the plume. Difficulties in making the measurements, however, preclude a firm statement as to the long-range radon daughter build-up during transport of the plume. It may well be that over distances of a few kilometers and time periods of an hour or more, that the radon daughters do approach equilibrium with their radon parent which has come from the mine exhaust vents.

To determine if this is actually true would require some additional measurements. We recommend that as time and funding permit, a significant research program be initiated to determine the ratio of radon to its daughters at considerable distances from mine exhausts or stockpiles and tailings piles in the uranium mining and milling areas. This information

is needed to determine in an absolute way what the local and regional effect of radon release from mine ventilation air and from other sources is.

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[1] "Measurement of Radon Daughters in Air by Alpha Counting of Air Filters," Jess W. Thomas. HASL-256, U. S. Atomic Energy Commission, April 1972.

[2] "Modification of the Tsivoglov Method for Radon Daughters in Air," Jess W. Thomas. Health Physics 19:691, November 1970.

TABLE 1. RADON AND DAUGHTER RATIOS AND CONCENTRATIONS AS A FUNCTION OF DISTANCE DOWNWIND FROM A DRY UNDERGROUND URANIUM MINE VERTICAL VENT

CONCENTRATIONS (pCi/liter)

Measurement/ Sample Site	Vent Exhaust	30*	30*	85*	140*
Rn	3400 ± 160***	0.64 ± 0.17	1.14 ± 0.17	6.74 ± 0.32	0.69 ± 0.12
RaA	1100 ± 35	<0.6	0.48 ± 0.40	2.22 ± 1.48	0.37 ± 0.47
RaB	726 ± 49	0.037 ± 0.13	0.29 ± 0.12	1.54 ± 0.77	0.11 ± 0.25
RaC	702 ± 10	0.18 ± 0.10	0.24 ± 0.11	2.37 ± 0.50	0.16 ± 0.16
Working Levels	7.43 ± 0.25	0.0006 ± 0.0008	0.0029 ± 0.0008	0.0189 ± 0.0046	0.0015 ± 0.0015

DAUGHTER/RADON RATIOS

Distance from Vent	RaA/Rn	RaB/Rn	RaC/Rn	Wind (m/sec)
Vent exhaust	0.32 ± 0.02	0.21 0.02	0.21 0.01	5
30* A**	<1.0	0.06 0.21	0.28 0.18	2
30* B	0.42 ± 0.36	0.25 0.11	0.21 0.21	2
85*	0.33 ± 0.22	0.23 0.11	0.35 0.08	2.5
140*	0.54 ± 0.70	0.16 0.37	0.23 0.24	1.5

* Indicates meters
** Wind direction changed early in run. Resampled after wind direction returned to original.
*** 3800 pCi/ℓ from Eberline radon monitor data.

TABLE 2. RADON AND DAUGHTER RATIOS AND CONCENTRATIONS AS A FUNCTION OF DISTANCE DOWNWIND FROM A WET UNDERGROUND URANIUM MINE HORIZONTAL VENT

CONCENTRATIONS (pCi/liter)

Measurement/ Sample Sets	Vent Exhaust	30*	96*	283*
Rn	616 ± 59**	32.4 ± 1.6	0.28 ± 0.09	0.29 ± 0.09
RaA	139 ± 9	3.0 ± 1.3	<0.6	0.07 ± 0.31
RaB	135 ± 5	0.89 ± 0.79	0.01 ± 0.16	0.04 ± 0.12
RaC	120 ± 3	1.33 ± 0.48	0.14 ± 0.11	0.08 ± 0.10
Working Levels	1.28 ± 0.03	0.013 ± 0.005	0.0004 ± 0.0009	0.0006 ± 0.0008

DAUGHTER/RADON RATIOS

Distance from Vent	RaA/Rn	RaB/Rn	RaC/Rn	Wind (m/sec)
Vent Exhaust	0.23 ± 0.03	0.22 ± 0.02	0.20 ± 0.02	---
30*	0.092 ± 0.042	0.027 ± 0.024	0.041 ± 0.015	2.7
96*	<2.3	0.04 ± 0.60	0.50 ± 0.44	1.3
283*	0.24 ± 1.11	0.14 ± 0.45	0.27 ± 0.36	2.3

* Indicates meters
** 563 pCi/ℓ on Eberline constant radon monitor

TASK 3

INCENTIVES FOR USING ELEVATED RELEASE POINTS TO REDUCE GROUND LEVEL RADON CONCENTRATIONS FROM UNDERGROUND URANIUM MINES

INTRODUCTION

Underground uranium mine ventilation air is generally exhausted at ground level. These ground-level releases result in higher radon concentrations at distances nearer the source than would result from releases from an elevated point. The advantage of elevated releases in terms of ground-level concentration diminish with distance from the source. This study models the long-term effects (on ground-level concentrations) of release height, of atmospheric mixing heights, of direction and distance from source, and of several source configurations for "typical" joint frequency distributions of wind speed, wind direction, and atmospheric stability.

EXPERIMENTAL PROTOCOL

The models most frequently used to describe downwind concentrations resulting from a short-period plume release are based on the assumption of a Gaussian plume spread in both the vertical and horizontal. For relatively long periods of time (say a month or longer), it is reasonable to assume that the wind direction within a selected sector of a 16-sector compass is randomly distributed, and thus it can be assumed that an effluent is uniformly distributed in the horizontal within the sector. Turner[1] presents appropriate Gaussian model equations for average ground-level concentration which are of the form

$$\frac{\overline{X}}{Q} = 2.03 \left(\frac{1}{\sigma_z \bar{u} x}\right) \exp\left[-0.5\left(\frac{H}{\sigma_z}\right)^2\right] \qquad (1)$$

or

$$\frac{\overline{X}}{Q} = 2.55 \left(\frac{1}{L \bar{u} x}\right) \qquad (2)$$

depending on whether a stable layer aloft is affecting the vertical distribution. In this notation, $\overline{X}$ is concentration, Q is pollutant emission rate (assumed uniform), σ_z is the standard deviation of pollutant concentration

in the vertical, $\bar{u}$ is the mean wind speed affecting the plume, x is downwind distance, H is stack (or plume) height, and L is the height of the stable layer which limits vertical plume mixing.

At distances far removed from the source, the exponential term in equation (1) approaches unity as σ_z becomes large compared to H. When a stable "lid" caps vertical dispersion, the vertical distribution of pollutant finally becomes uniform, σ_z can be equated to L, and equation (1) becomes identical in form to equation (2).

The point to be made for the subject task is that at a selected large downwind distance x, σ_z is generally considered to be identical for both ground level and elevated releases. (Also, L is the same whether equation (2) is applied to a ground-level or an elevated release.) Hence, $\bar{u}$ is the only variable in equations (1) and (2) that can call for a difference in normalized concentration χ/Q at distances far removed from the source.

Most models use the wind speed measured at (or extrapolated to) the release height. For instance, for a ground-level release, the wind speed measured near the surface source is input to equations (1) or (2), irrespective of the distance downwind for which χ/Q is calculated. Similarly, the (greater) wind speed measured at stack level would be input to prescribe χ/Q from a stack, again irrespective of downwind distance. One is left with the intuitively unappealing result that at, say, 500 km downwind of a pair of sources (one at ground level, the other from a stack), the concentration resulting from the stack would be less than that resulting from the surface release. At such a great distance, the source height wind speed difference should not be effective.

A technique has been developed to permit a more realistic approach to the wind speed effective in transporting pollutants. The effective wind height is presumed to grow with distance so as to better apply to the entire vertical distribution being transported. For a ground-level source, the effective wind height, E, is presumed equal to $0.5\ \sigma_z$. If a stable layer caps the mixing layer, $E = 0.5\ L$ after σ_z equals L. For an elevated release, the following relationships are presumed:

If $\sigma_z \leq H$, $E = H$

If $\sigma_z > H$ and $\sigma_z \leq L\text{-}H$, $E = 0.5(\sigma_z + H)$

If $\sigma_z > H$ and $\sigma_z > L\text{-}H$, $E = 0.5\ L$

The wind speed is extrapolated from measurement height (anemometer height) to height E by means of a technique presented by Budney[2]. The vertical dispersion parameter, σ_z, is calculated as a function of travel distance[3]. The calculated values of σ_z closely match the widely used values presented by Gifford[4] in graphical form.

DISCUSSION

In order to apply the selected diffusion and transport model, it was necessary to have meteorological data available in the form of joint frequency distributions of wind speed, wind direction, and atmospheric stability for annual periods. A relatively large number of such annual joint frequency distributions have been generated in the process of meeting licensing requirements associated with nuclear power plants.

From this large number, six sets of joint frequency distributions were chosen for more careful examination. The sites of these meteorological measurements were Hanford (southeast Washington), Black Fox (Tulsa, Oklahoma), Robinson (Hartsville, South Carolina), Rancho Seco (Sacramento County, California), Surrey (Surrey County, Virginia), and Trojan (Columbia County, Oregon) plants. Hanford was chosen partially because it is in a dry, near-desert climate, a climate similar to western New Mexico. The other data sets were chosen randomly to get some idea of the range of possible wind speed and stability distributions.

Plots of cumulative distributions of wind speed at an elevation of 10 meters revealed that Hanford displayed a representative median distribution for speeds through about 5 m/sec (11 mph). The Black Fox distribution showed the lowest cumulation frequency of winds at low wind speeds. Distributions of atmospheric stability revealed that Hanford was also reasonably representative of the "average" site. On the other hand, Black Fox was the site of the highest frequency of stable atmospheres.

The scope and funding of this task did not permit an examination of concentrations resulting from a large number of the possible combinations of site meteorology, source configurations, source heights, and atmospheric mixing heights. Site meteorology (joint frequency distribution of wind speed, wind direction, and atmospheric stability) was arbitrarily limited to two sites--Hanford and Black Fox. Hanford was selected because of its "average" meteorology and, as stated earlier, because it has a similar climate to western New Mexico. Black Fox was chosen primarily because of its bias toward stable atmospheres--a meteorological condition which should maximize the effects of different source heights. Hanford was also found to be the most similar to western New Mexico in terms of mean annual mixing height than any of the other five candidate meteorological sites.[5]

All concentrations reported in this study are annual average values.

Results

Figures 1(a) and 1(b) give the modeled areal distribution of annual average χ/Q at ground level* which would result from a constant-rate surface release. A 200-m mixing height and Hanford meteorology were used. Figure 1(a) gives the concentration within 5 miles of the release point. Figure 1(b) gives isopleths of concentration to a radius of 50 miles of the source. The area with χ/Q between 1×10^{-7} and 2×10^{-7} sec/m^3 is shaded on both parts of Figure 1 as an aid to perceiving the scale relationship. Figures 2(a) and 2(b) give the same data for the Black Fox site.

Note that the areas under comparable isopleths on Figures 1(a), 1(b), 2(a), and 2(b) are reasonably similar in area despite the differences in shape. Recall that the meteorology at Hanford was deemed "average" while Black Fox had relatively high wind speeds (which reduce downwind concentrations) and relatively stable atmospheric conditions (which increase downwind concentrations). The net effect is that despite the difference in meteorology, the overall effect of dosage on an evenly distributed population should be quite similar for the ground-level release case.

The joint frequency distributions of wind speed, wind direction, and atmospheric stability from which the Figures 1(a), 1(b), 2(a), and 2(b) data were generated listed wind directions for sixteen 22-1/2 degree sectors

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* All concentrations and concentration ratios presented in this study pertain to ground level irrespective of effluent release height.

(i.e., for north, north-northeast, northeast, etc.). A sum of all χ/Q values at a given distance from the source is proportional to the mean concentration passing that radial distance. Curves depicting the loci of this along arc (crosswind) sums of concentration are given in Figure 3. Although mixing height used in these computations was 1000 m [as opposed to the 200 m used in Figures 1(a), 1(b), 2(a), and 2(b)], it is evident again that Hanford and Black Fox generate quite similar magnitudes of concentration despite the difference in input meteorology. As before, a surface release was modeled.

Curves similar to those presented in Figure 3 can be generated for elevated releases. Release heights of 20 m, 50 m, and 100 m and a mixing height of 1000 m were modeled with the Hanford and Black Fox meteorology. Figure 4 presents the results in the form of ratios rather than sums of χ/Q. The numerator in a plotted ratio curve is the sum of χ/Q from an elevated release, while the denominator is the sum of χ/Q from the appropriate ground-level release as depicted in Figure 3.

Figure 4 shows that despite the strong similarity of concentration resulting from ground-level releases at Hanford and Black Fox (difference of only 7% at 0.75 miles increasing to 24% at 45 miles), there are distinct differences resulting from elevated releases. An examination of the data reveals that the ground-level concentrations resulting from a 100-m stack ($H = 100$ m) at Hanford is roughly twice the concentration that would result from a 100-m stack at Black Fox, irrespective of the distance from the source. The difference between Hanford and Black Fox downwind, summed concentrations is not as dramatic for the 50-m or the 20-m release height (especially at the larger distances), but is still appreciable. For instance, at 0.75 miles where the concentration resulting from a Hanford ground-level release is 7% less than at Black Fox (Figure 3), a release from an elevation of 20 m at Hanford results in a ground-level summed concentration 33% greater than at Black Fox (Figure 4).

An elevated release is more effective with Black Fox meteorology because of the higher frequency of stable atmospheres. The effects of an elevated release are propagated to greater distances under stable conditions.

Figure 4 also gives an indication of the general magnitude that annual average concentration can be reduced by increasing source height to 20 m,

50 m, or 100 m. Of course, the benefits of an elevated release are greater for a greater release height and for distances close to the source. The 20-m release height reduces ground-level concentrations by 50 to 70% at distances near a mile of the source, but by only 30% at a distance of 10 miles.

The expected benefits of lower concentration which accrue as a result of increased release height can be presented in the form of areal plots of concentration ratios. Figures 5(a), 5(b), 6(a), 6(b), 7(a), and 7(b) present data following input of Black Fox meteorology and modeled release heights of 20 m, 50 m, and 100 m, respectively. Figures 5(a), 6(a), and 7(a) present the concentration within 5 miles of the release point, while Figures 5(b), 6(b), and 7(b) give the isopleths of concentration to a radius of 50 miles from the source. A mixing height of 200 m was input in all cases. The isopleths presented in these figures result from ratios generated for 320 specific points (16 directions at 20 distances). Each individual ratio resulted from the modeled elevated-release concentration divided by the modeled ground-level-release concentration for the same location. Ratios were converted to percentages by multiplying by 100. Thus, an isopleth labeled 50% indicates that the elevated release reduced the annual average ground-level concentration to half the value computed for a ground-level release.

Although the isopleths in Figures 5, 6, and 7 are labeled, these figures probably serve better as qualitative representations of the effects of increasing release height than they do in quantifying those effects. Note that the major axes of the isopleths (representing greater stack height benefits at a given distance) lie east-west, whereas the major axis of concentration resulting from a ground-level release [Figures 2(a) and 2(b)] lies in a north-south direction. Thus, the greatest percentage reduction in concentration from elevated releases is <u>not</u> in the areas where it would do the most good at Black Fox. Although the data are not shown, the axes of isopleths for Hanford [similar to those shown in Figures 5(a), 5(b), 6(a), 6(b), 7(a), and 7(b) for Black Fox] <u>do</u> align with the axis of maximum concentration shown in Figures 1(a) and 1(b). Thus, at Hanford an increase in release height is more effective in reducing concentrations in directions of higher concentration than in directions of lower concentration. On the

basis of the conflicting Hanford and Black Fox data, it appears that the axis of maximum benefit from elevated release cannot be forecast on the basis of ground-release concentration pattern.

Figure 8 denotes that an elevated release can change the direction at which a maximum concentration is observed. With a ground-level release at Black Fox, the maximum concentration was observed at a direction north-northwest of the source at all distances. However, the modeled 20-m release resulted in higher concentrations in a direction due north of the source for distances closer than one mile. At distances beyond a mile, the major axis of concentration remained north-northwest of the source irrespective of the source height.

Not surprisingly, the authors have concluded that the data generated in this limited study do not permit generation of hard and fast general rules. Although more data and more intense analysis might help to better define the range of benefits that could accrue from elevated releases, it is felt that analysis of the effects of elevated releases is a very site-specific procedure. This statement is particularly applicable in terms of assessing the dosage expected to be received by receptors in the vicinity of a mine. But as with our other results, the Black Fox and Hanford data can offer some rough estimates of elevated release on total dosage impact.

By making the assumption that the population (or other receptors) are equally distributed within a given circle or a given annulus around a mine, a number proportional to total population (receptor) dosage can be computed. The procedure involves dividing the total area of concern into a number of smaller areas, multiplying each smaller area by the appropriate annual average concentration, and then summing all the products. If such assumed products are developed for elevated and for ground-level releases, a unitless ratio can be formed which compares the dosage from the elevated release to the dosage from the ground-level release. Table 1 presents these dosage ratios for two specific areas: 1) the entire area within 50 miles of the plant; and 2) the annular area between 0.5 and 5.0 miles from the plant. The table presents ratios based on both Hanford and Black Fox meteorology.

Table 1 shows that the influence of mixing height on the ratio is small--especially with the release heights of more practical concern (20 m

and 50 m). The largest practical difference is seen for the Hanford meteorology and the 0 to 50 mile area of concern. Here the 200-m "lid" dictates a ratio of 0.49 for the 50-m release while the 1000-m lid results in a ratio of 0.43 for the same release height.

It is rather pointless to discuss Table 1 in great detail. Suffice it to say that for the 0.5 to 5.0 mile annulus and for the release heights of practical concern (ground level, 20 m, and 50 m), an increase in release height from ground level to 20 m roughly halves the dosage, and an increase from 20 m to 50 m again roughly halves the dosage. Note also that an increase in release height is more effective with the more stable Black Fox meteorology than with the Hanford meteorology.

Graphical presentation of the data in Table 1 permits the estimation of the effects of stack releases other than the specific heights tabled. Figure 9 graphically presents these data. This figure also illustrates that the benefits in reduced dosage per unit of increased stack height decrease with increasing stack height.

Figures 10(a) through 10(g) present areal distributions of concentration rations (χ/Q for 20-m release/χ/Q for ground-level release) for a series of source configurations. The meteorological input was Black Fox, and the mixing height was presumed to be 200 m in all cases. The emission rate (Q) is presumed to sum to unity for all configurations. For example, if two sources are considered, each source is considered to emit at a rate Q/2. [An exception is Figure 10(g) where the east source is presumed to be 2/3 Q and the west source is presumed to be 1/3 Q.) The diameter of the circle portrayed on each figure is 5 miles. Sources are deployed in east-west, north-south, or equilateral triangular configurations. With the exception of Figure 1(a) (where only a single source is considered), the spacing between sources is one mile.

Funding and time restrictions did not permit a careful quantitative evaluation of the data presented in Figures 10(a) to 10(g). Nonetheless, some qualitative evaluation of these figures is instructive. The alignment of the sources does stretch the axes of the ratio isopleths along a similar axis. And although the ratios in the immediate vicinity of the source(s)

TABLE 1. Dosage Ratios Based on Hanford and Black Fox Meteorology. Ratios Are Elevated Release Data/Ground-Level Release Data. Equal Distribution of Receptors Assumed.

Area of Concern	Source of Meteorology	Mixing Height: 200 m, Release Height: 20 m	200 m, 50 m	200 m, 100 m	1000 m, 20 m	1000 m, 50 m	1000 m, 100 m
Circle, 0 to 50 Miles	Black Fox	0.59	0.37	0.19	0.58	0.34	0.15
	Hanford	0.66	0.49	0.35	0.63	0.43	0.25
Annulus, 0.5 to 5.0 Miles	Black Fox	0.50	0.19	0.060	0.50	0.18	0.053
	Hanford	0.60	0.30	0.11	0.59	0.28	0.089

are higher with two sources than with one (or with three sources than with two), the total area embraced by the 40% isopleth does not vary much from one configuration to the next. One can observe that beyond the 40% isopleth (i.e., beyond about 1.5 miles from the centroid of sources), the effect of source configuration is not great. At a distance of 2.5 miles from the source, the 55% and 60% isopleths are most prevalent. Within 1.5 miles of the centroid of the sources, the multiple sources do flatten the ratio gradient and do eliminate the modeled ratio of zero in evidence at the single source stack base. Hence, at very near source centroid locations, the multiple elevated source configuration can be considered less desirable than a single source configuration.

The evidence in Figures 10(a) to 10(g) can be summarized by the statement that beyond 1.5 miles from the centroid of up to three 20-m stacks (spaced a mile or less apart), the effect of varying stack locations is negligible. It should be recalled, however, that for a specific site with specific meteorology and specific population distributions, even such minor differences as shown in Figures 10(a) to 10(g) could be important.

SUMMARY

The conclusions drawn are based on a reasonably small data base. Although some general guidance relative to the effects of elevated releases is offered, more definitive information could be generated on a site-specific basis, particularly in the case of strongly biased distributions of wind direction and/or population. Conclusions drawn with the aid of the Hanford and Black Fox data bases are:

- Despite differences in meteorology, different sites can have very similar annual average distributions of concentration.
- Despite similarity of magnitude and distribution of ground-level concentration resulting from ground-level releases, the two sites displayed quite different ground-level concentrations following modeled elevated releases. A greater reduction in concentration was associated with the site with more stable atmosphere (Black Fox).

[2] L. J. Budney, Guidelines for Air Quality Maintenance, Planning and Analysis, Volume 10 (Revised): Procedures for Evaluating Air Quality Impact of New Stationary Sources. EPA-450/4-77-001, Research Triangle Park, NC 27711, 1977.

[3] E. C. Eimutis and M. G. Konicek, "Derivations of Continuous Functions for the Lateral and Vertical Atmospheric Dispersion Coefficients." Atm. Environ. 6:859-863, 1962.

[4] F. A. Gifford, Jr., "Use of Routine Meteorological Observations for Estimating Atmospheric Dispersion." Nucl. Safety, 2(4):47, 1961.

[5] G. C. Holzworth, Mixing Heights, Wind Speeds, and Potential for Urban Air Pollution Throughout the Contiguous United States. Office of Air Programs Publication AP-101, U. S. EPA, U. S. Government Printing Office, Washington, D. C. 20402, 1972.

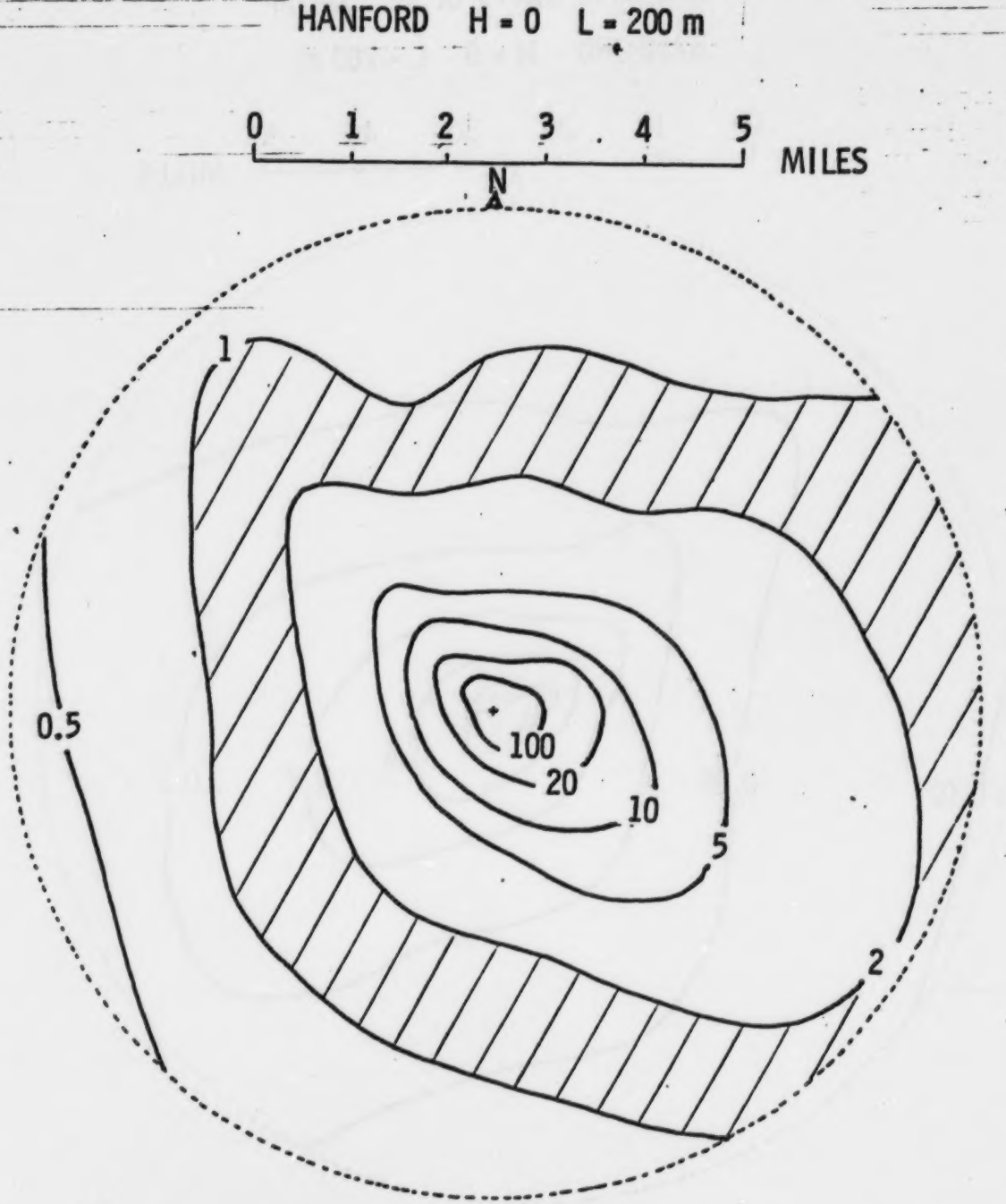


Figure 1a. Areal Distribution of χ/Q for Hanford Resulting from Surface Release. Mixing Height, 200m. Area Within 5 Miles of Source.

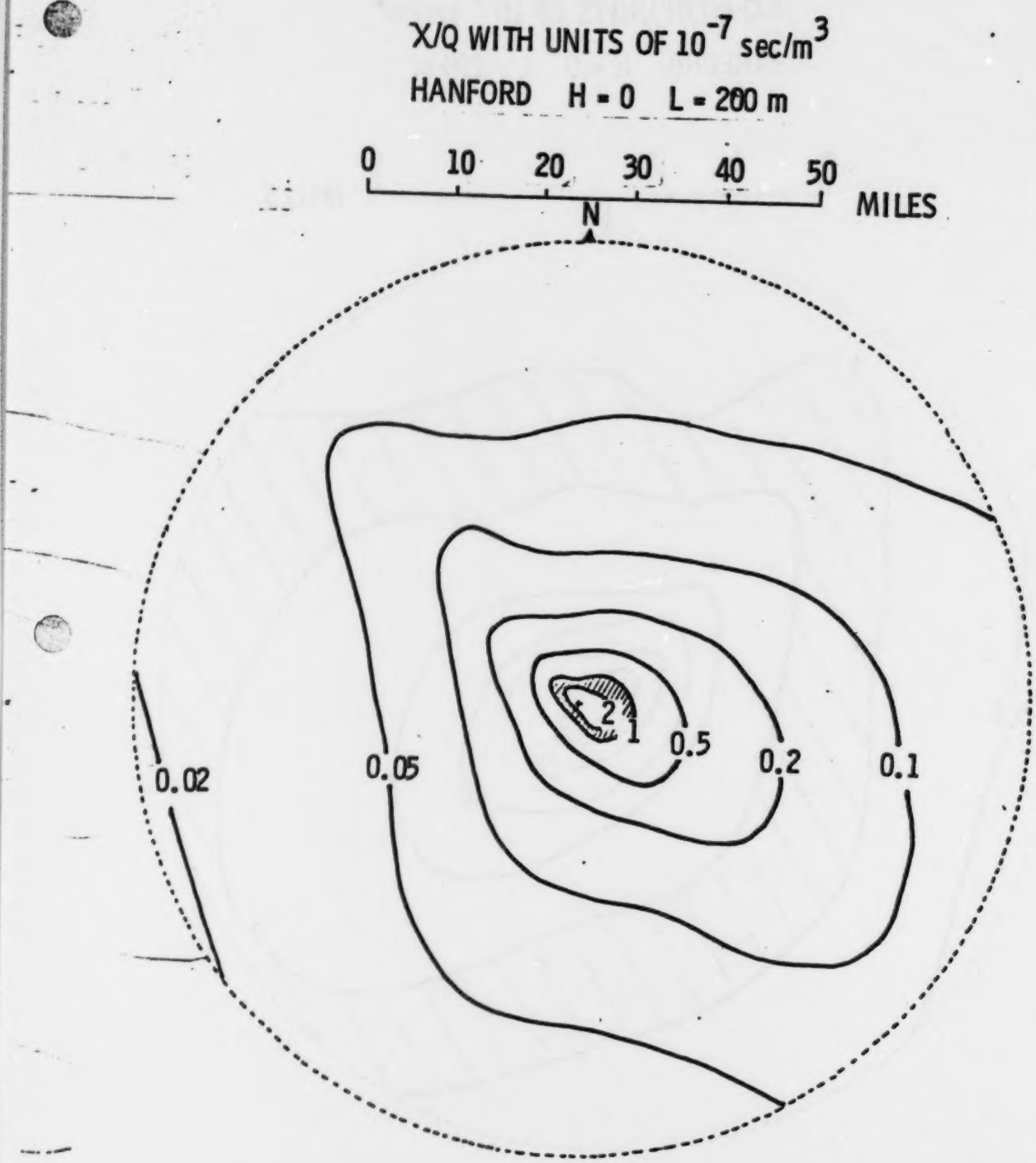


Figure 1b. Areal Distribution of χ/Q for Hanford Resulting from Surface Release. Mixing Height, 200m. Area Within 50 Miles of Source.

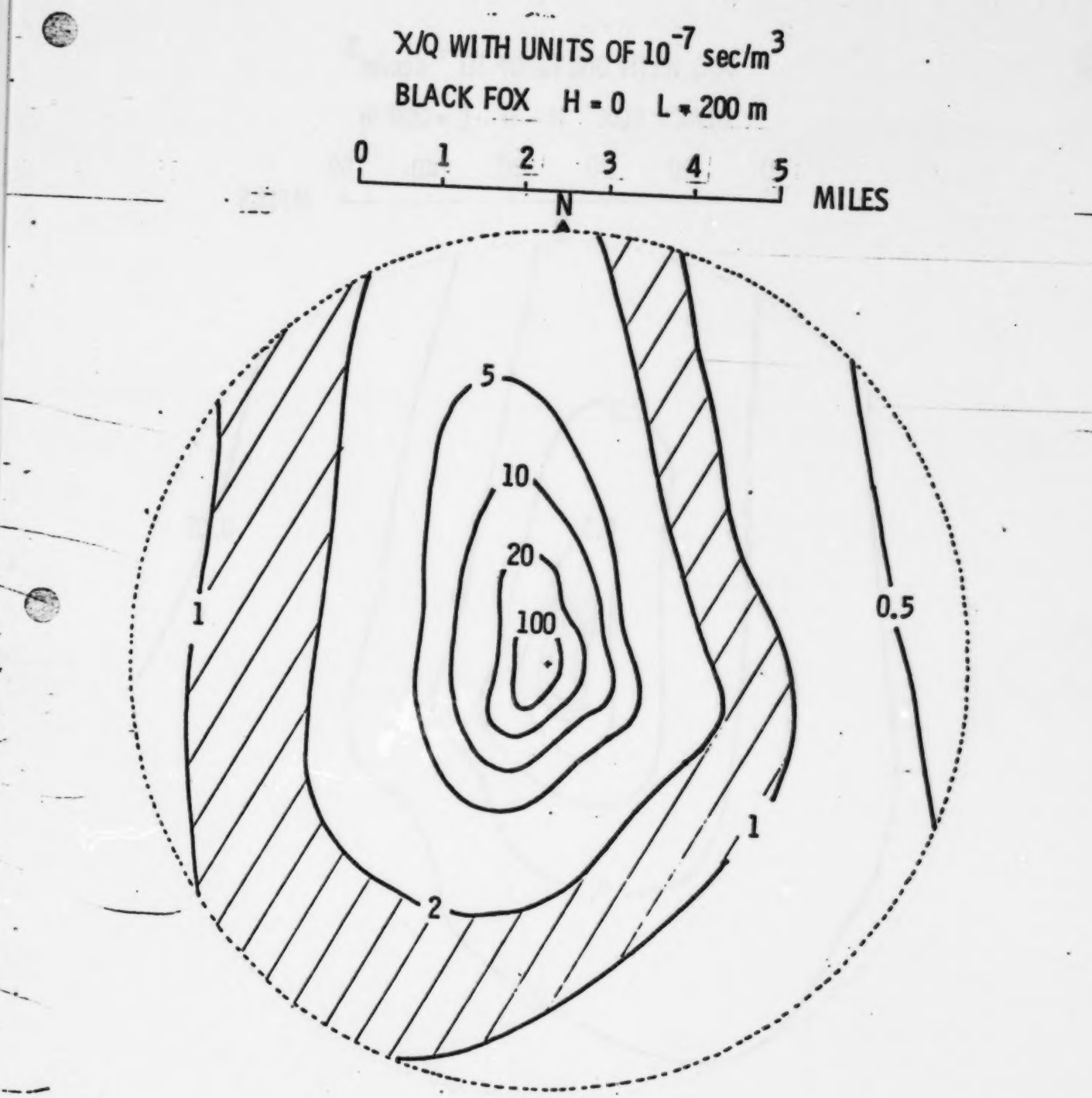


Figure 2a. Areal Distribution of x/Q for Black Fox Resulting From Surface Release. Mixing Height, 200m. Area Within 5 Miles of Source.

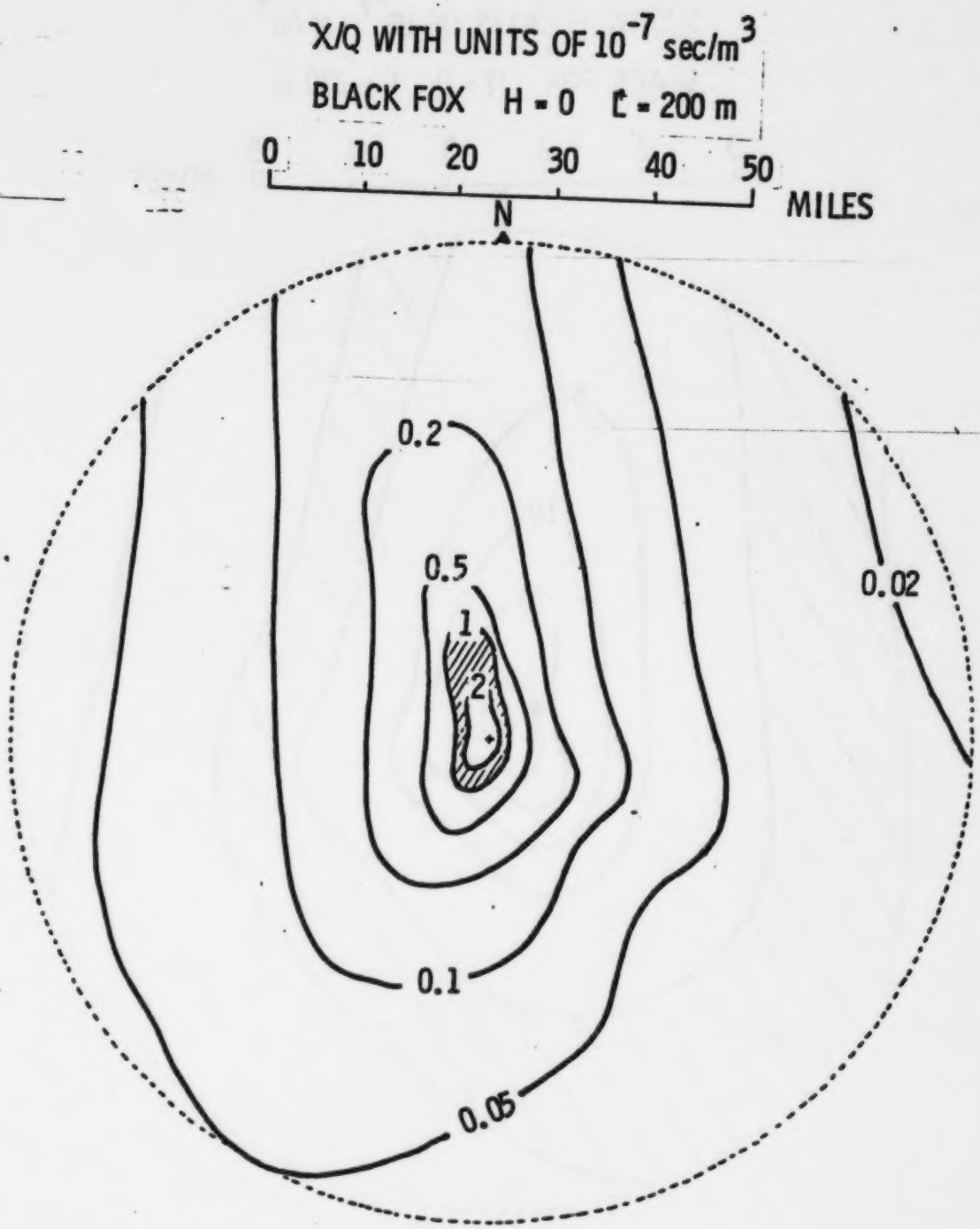


Figure 2b. Areal Distribution of χ/Q for Black Fox Resulting from Surface Release. Mixing Height, 200m. Area Within 50 Miles of Source.

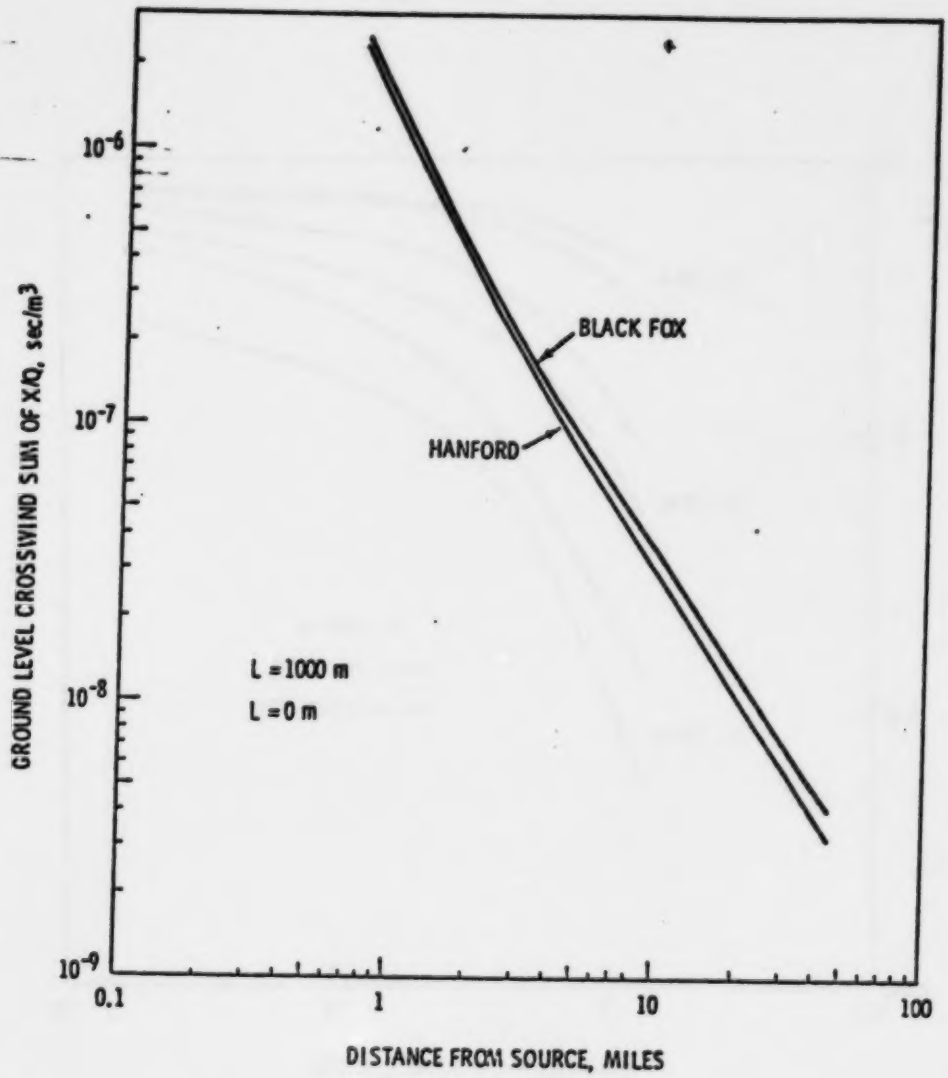


Figure 3. Crosswind Sum of Ground-Level χ/Q versus Distance from Source. Mixing Height of 1000m and Ground Level Release.

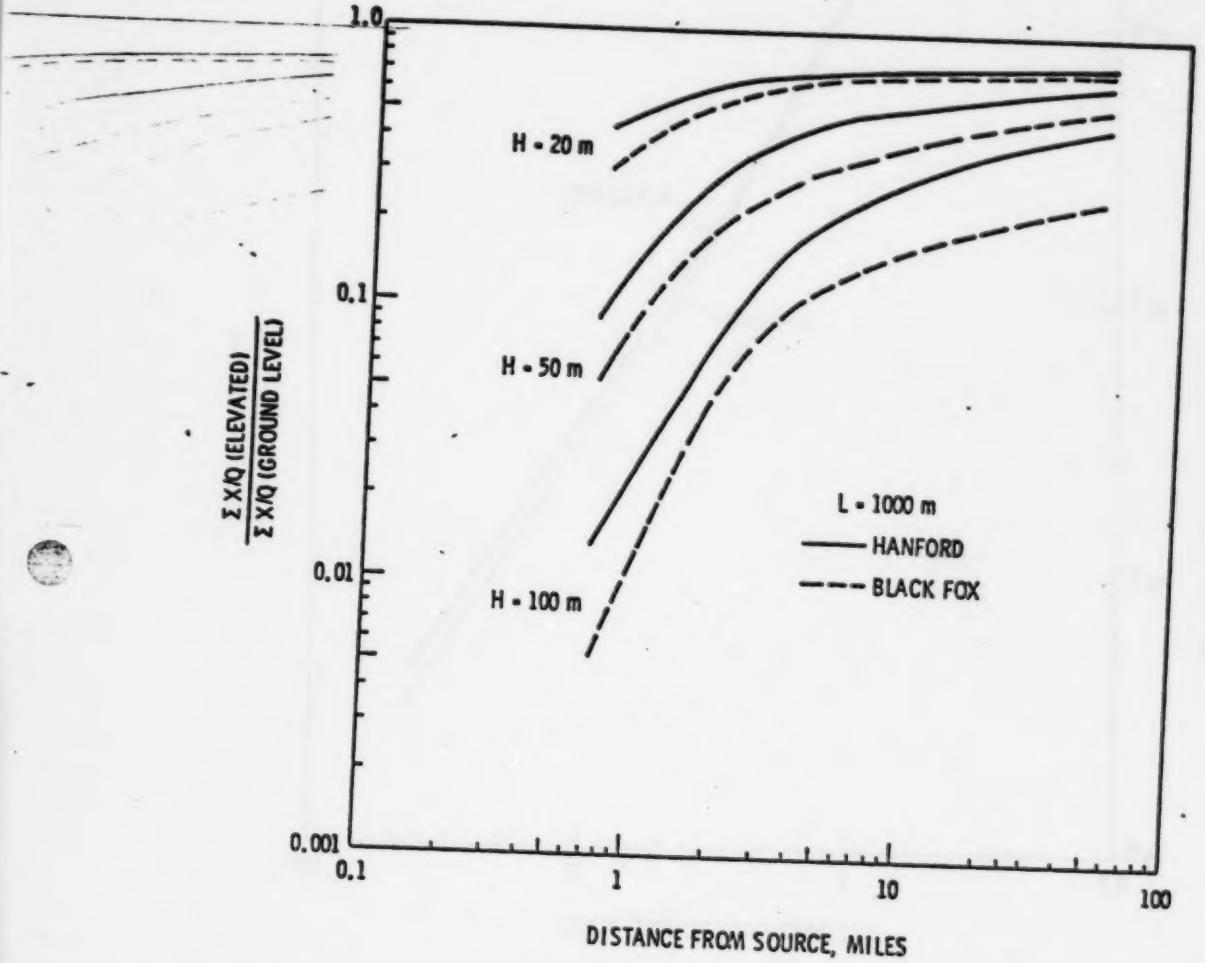


Figure 4. Ratios of Crosswind Sums of Ground Level x/Q versus Distance from Source. Mixing Height of 1000m.

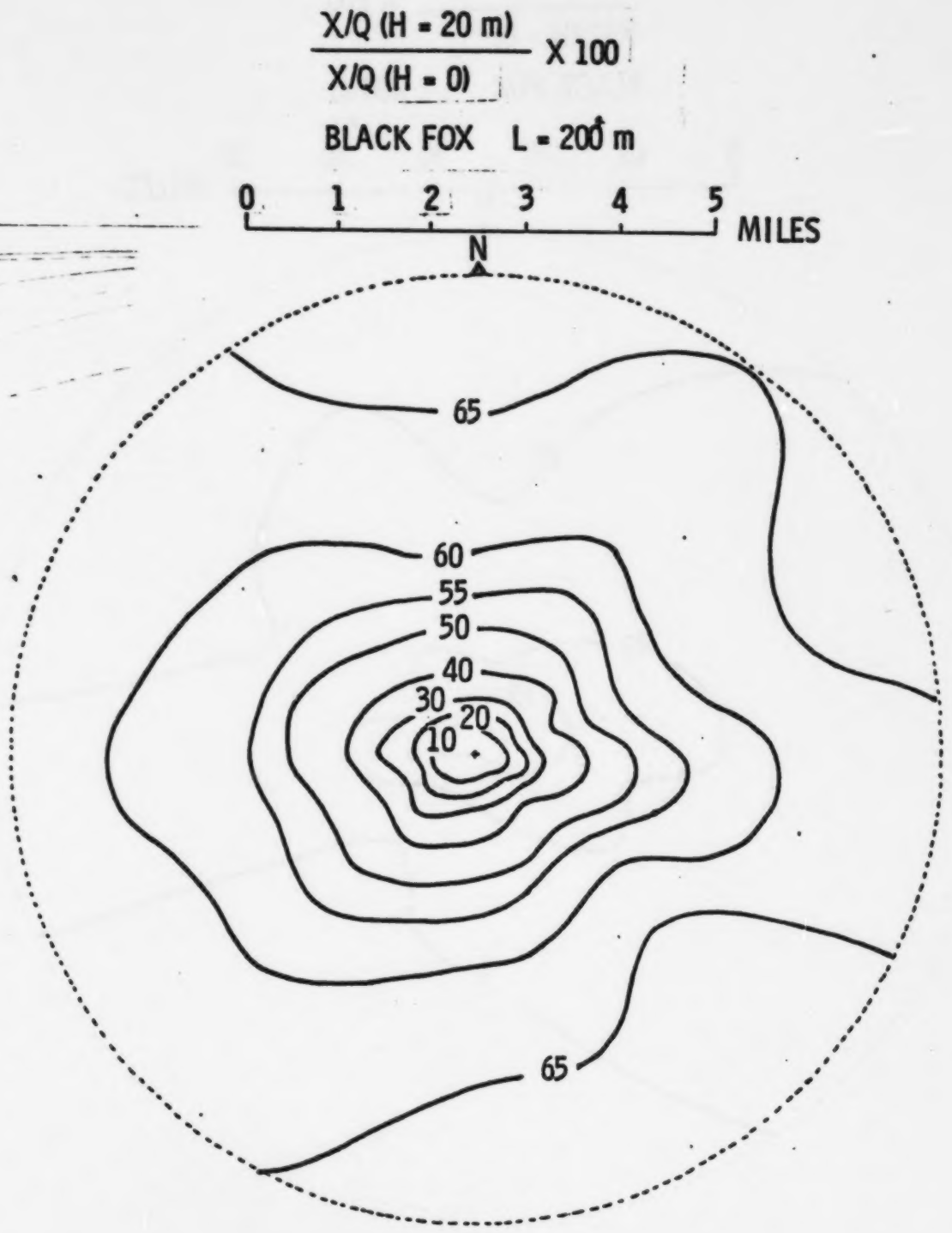


Figure 5a. Areal Distribution of Ratio of Ground Level χ/Q (H=20m) to Ground Level χ/Q (H=0) for Black Fox Meteorology. Mixing Height of 200 m. Area Within 5 Miles of Source.

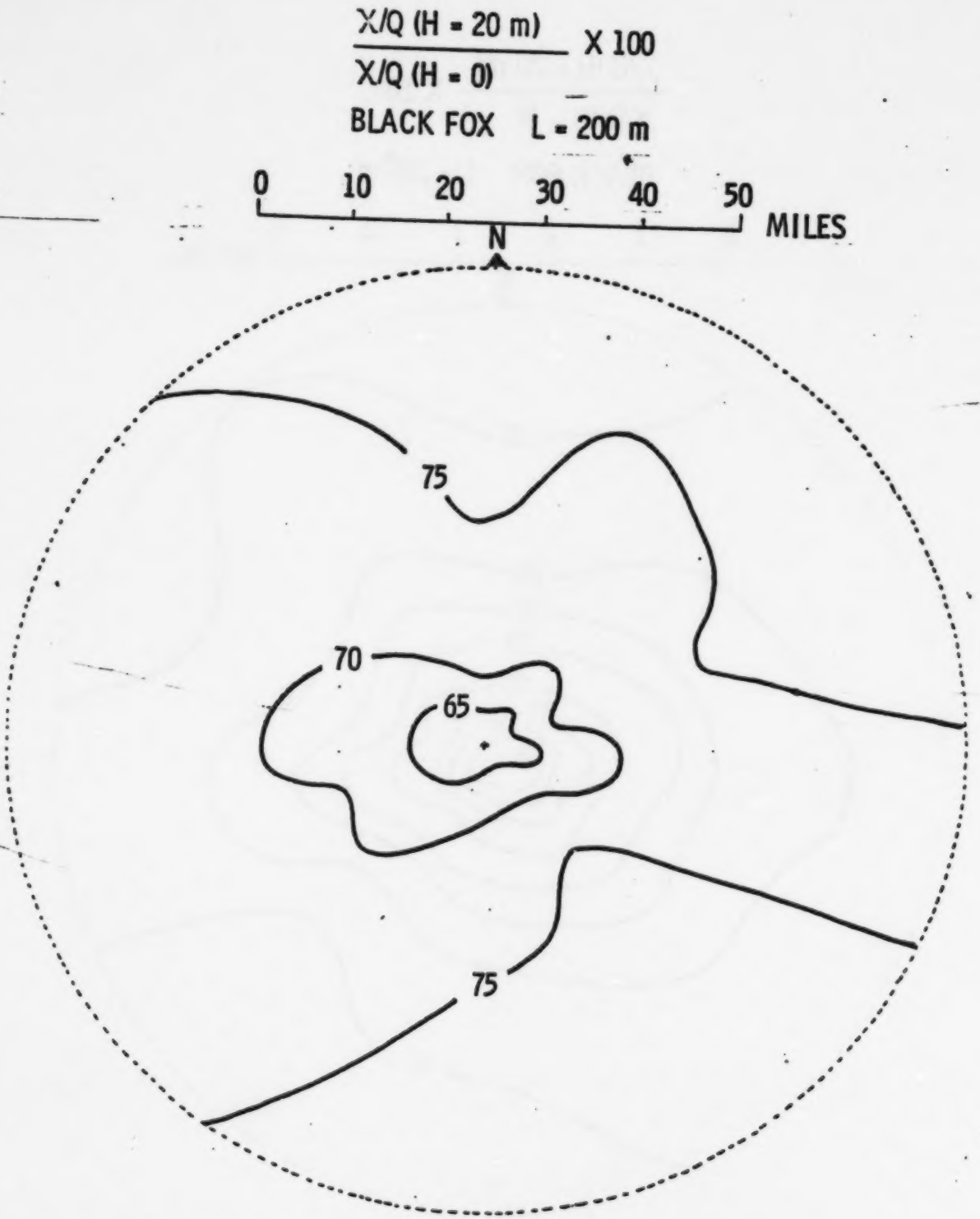


Figure 5b. Areal Distribution of Ratio of Ground Level x/Q (H=20m) to Ground Level x/Q (H=0) for Black Fox Meteorology. Mixing Height of 200m. Area With 50 Miles of Source.

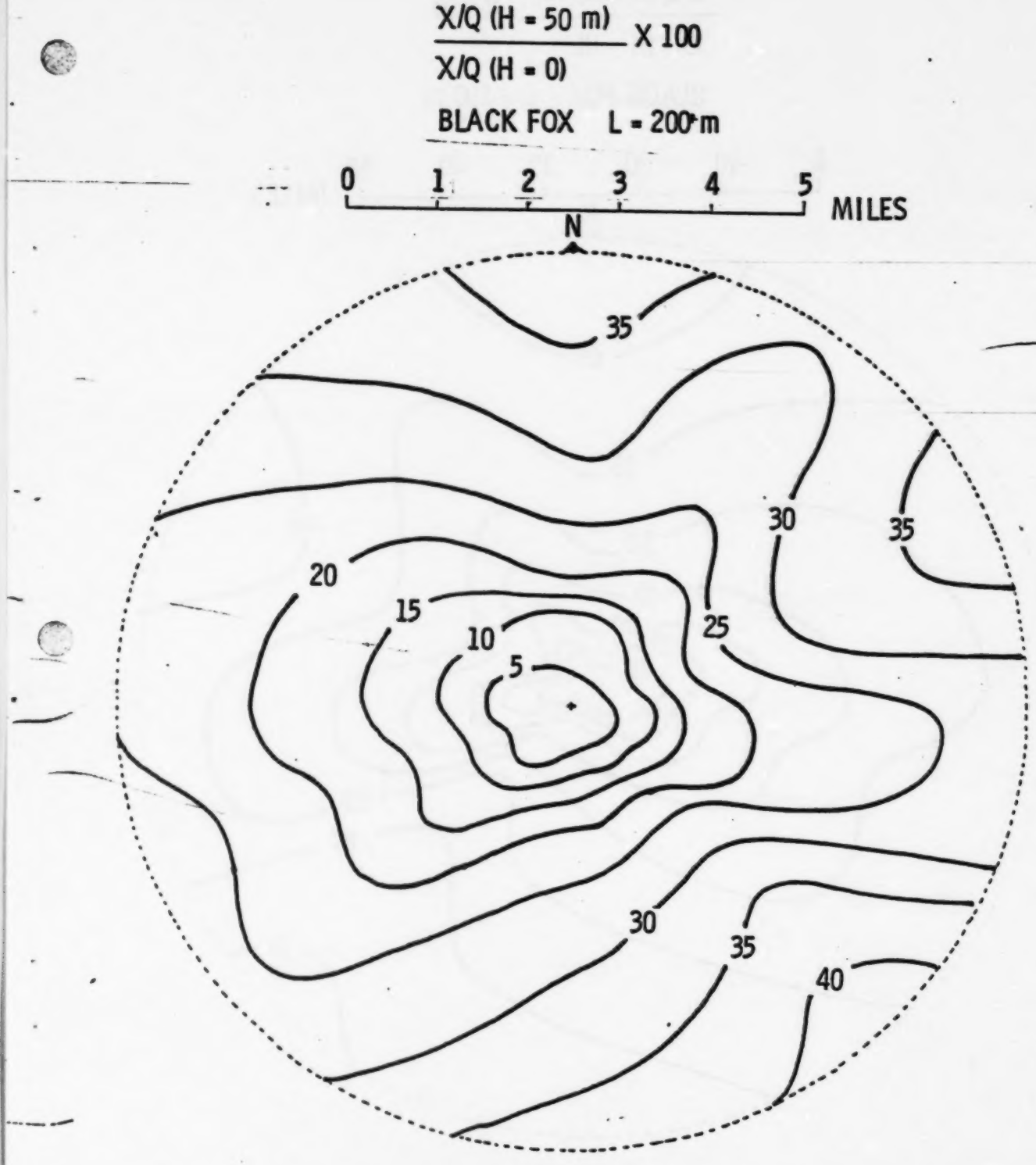


Figure 6a. Areal Distribution of Ratio of Ground Level χ/Q (H=50m) to Ground Level χ/Q (H=0) for Black Fox Meteorology. Mixing Height of 200m. Area Within 5 Miles of Source.

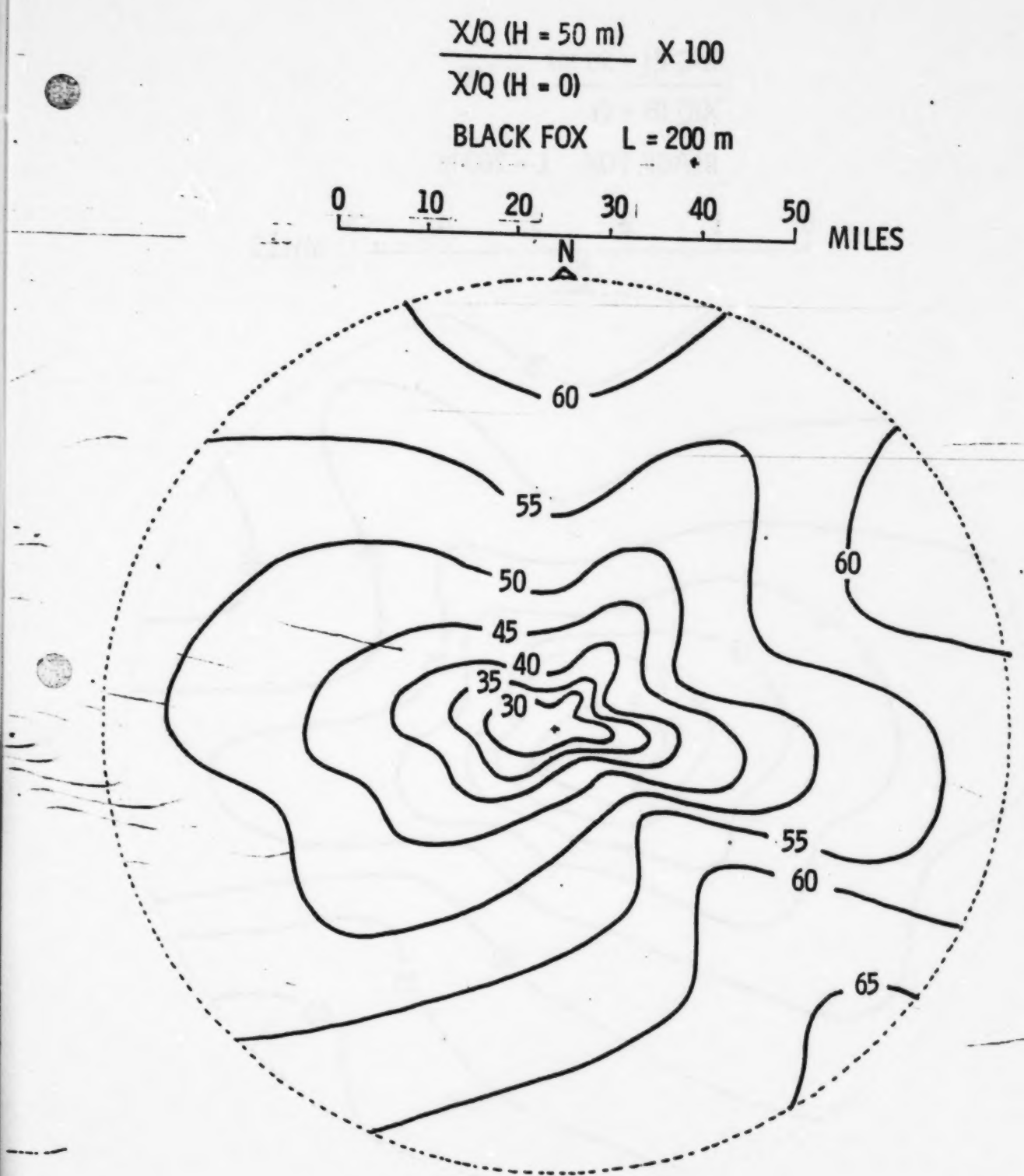


Figure 6b. Areal Distribution of Ratio of Ground Level x/Q (H=50m) to Ground Level x/Q (H=0) for Black Fox Meteorology. Mixing Height of 200m. Area Within 50 Miles of Source.

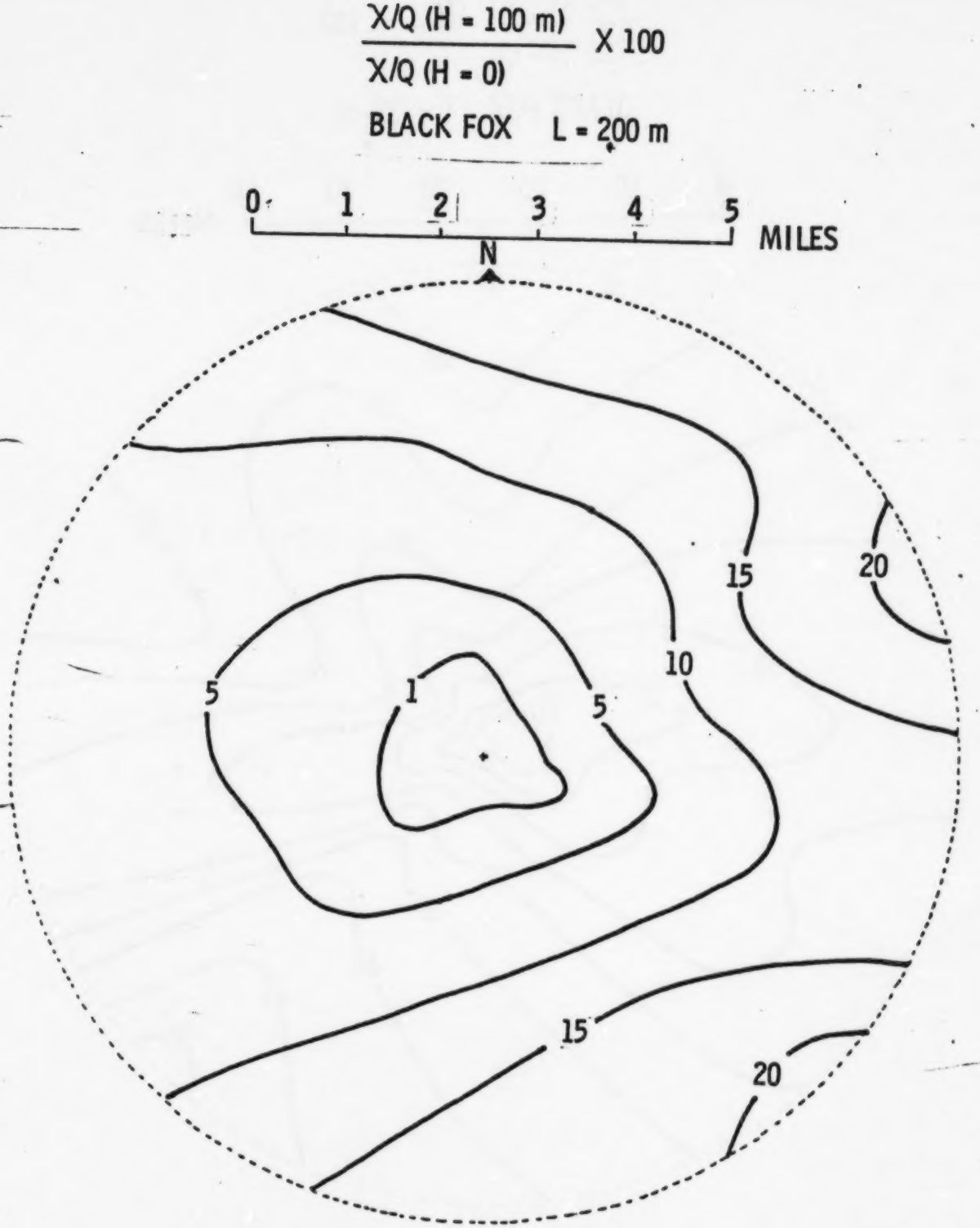


Figure 7a. Areal Distribution of Ratio of Ground Level χ/Q (H=100m) to Ground Level χ/Q (H=0) for Black Fox Meteorology. Mixing Height of 200m. Area Within 5 Miles of Source.

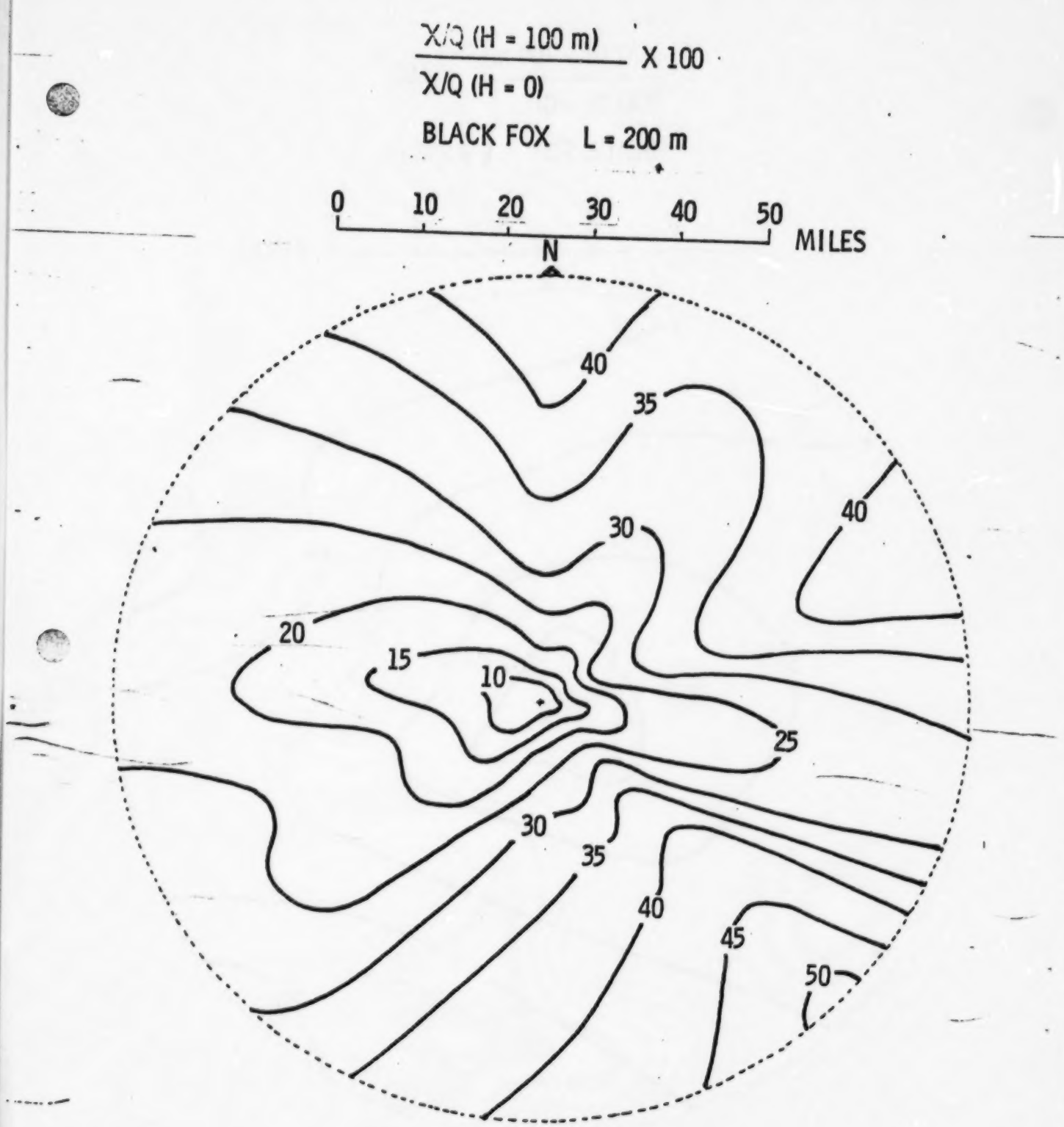


Figure 7b. Areal Distribution of Ratio of Ground Level χ/Q (H=100m) to Ground Level χ/Q (H=0) for Black Fox Meteorology. Mixing Height of 200m. Area Within 50 Miles of Source.

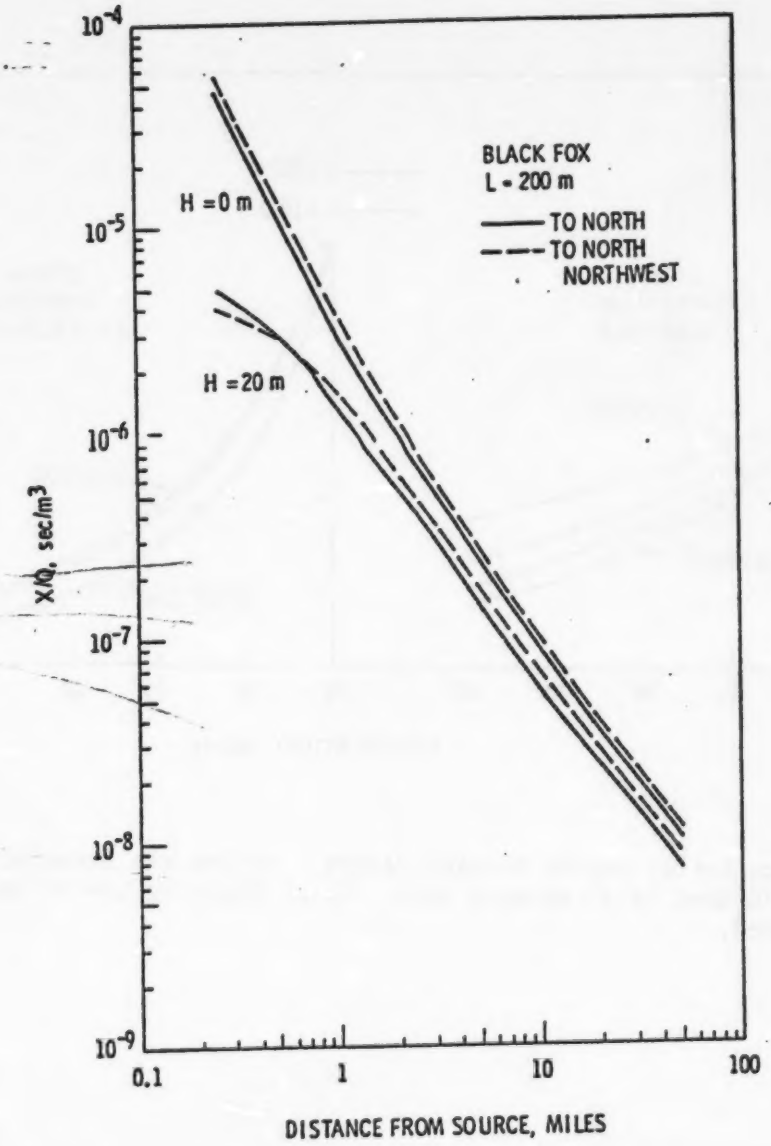


Figure 8. Ground Level χ/Q versus Distance from Source for Selected Directions and Release Heights at Black Fox. Mixing Height of 200m.

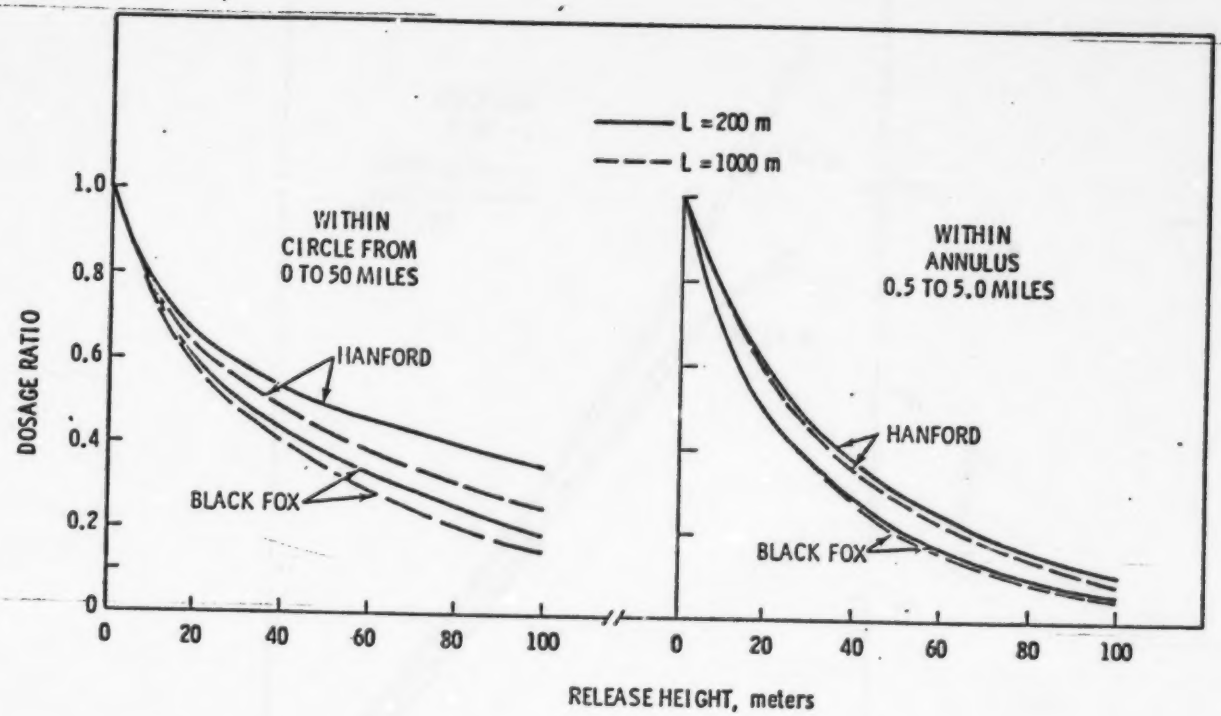


Figure 9. Dosage Ratios versus Release Height. Ratios are Elevated Release Data/Ground Level Release Data. Equal Distribution of Receptors Assumed.

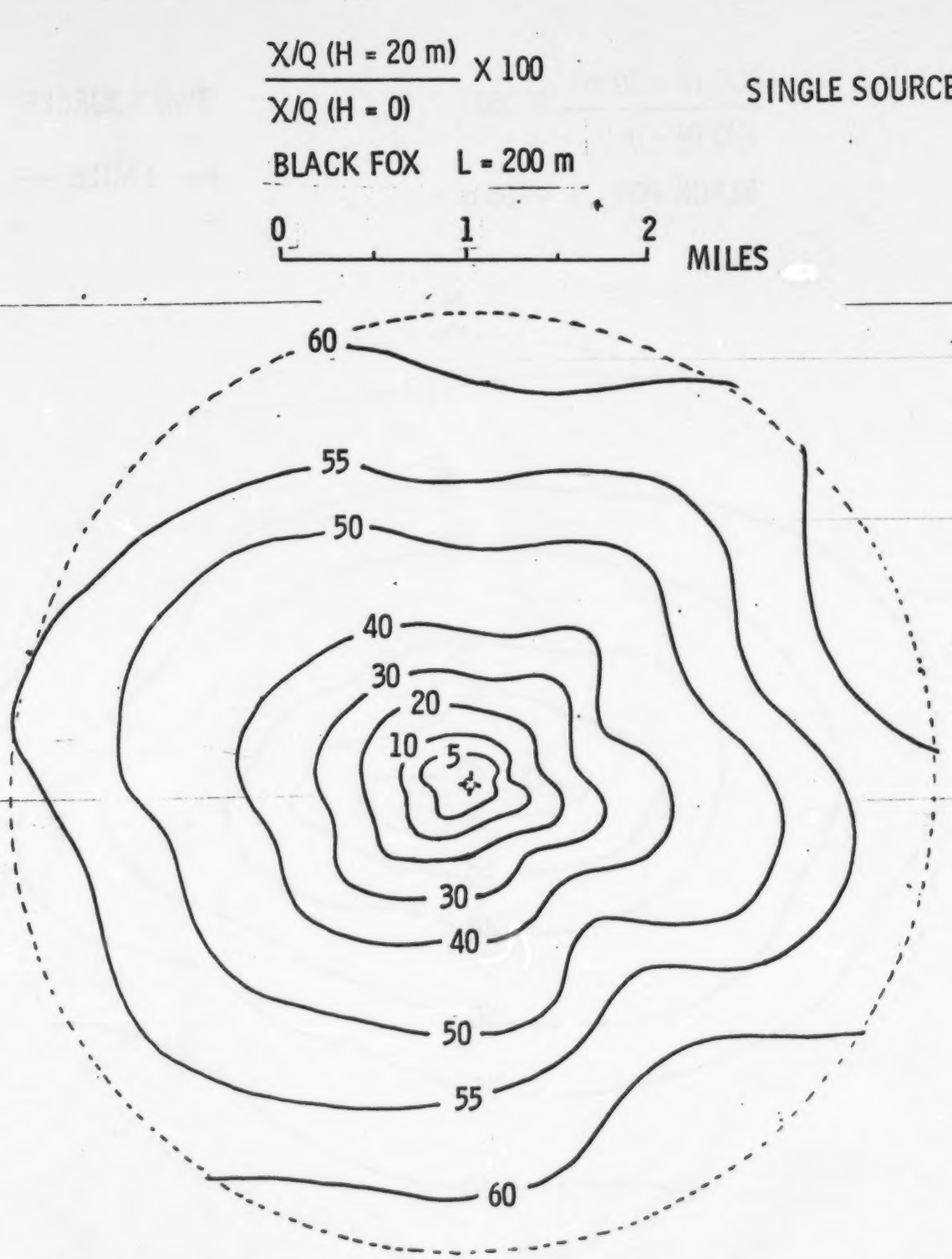


Figure 10a. Areal Distribution of Ratio of Ground Level χ/Q (H=20m) to Ground Level χ/Q (H=0) for Black Fox Meteorology. Mixing Height of 200m. Area Within 2 Miles of Black Fox. Single Source.

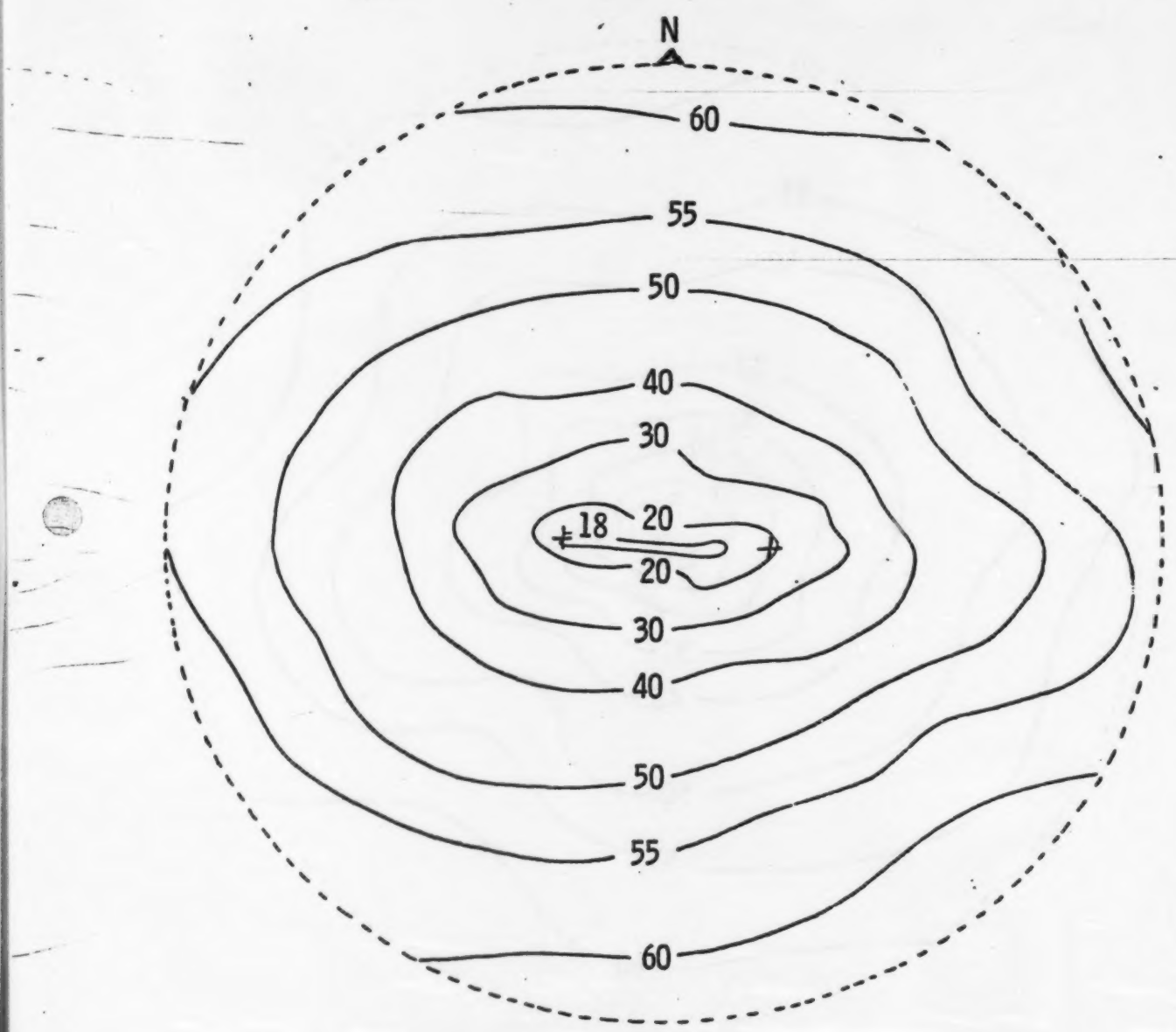


Figure 10b. Areal Distribution of Ratio of Ground Level χ/Q (H=20m) to Ground Level χ/Q (H=0) for Black Fox Meteorology. Mixing Height of 200m. Area Within 2 Miles of Black Fox. Depicted Dual Source Configuration.

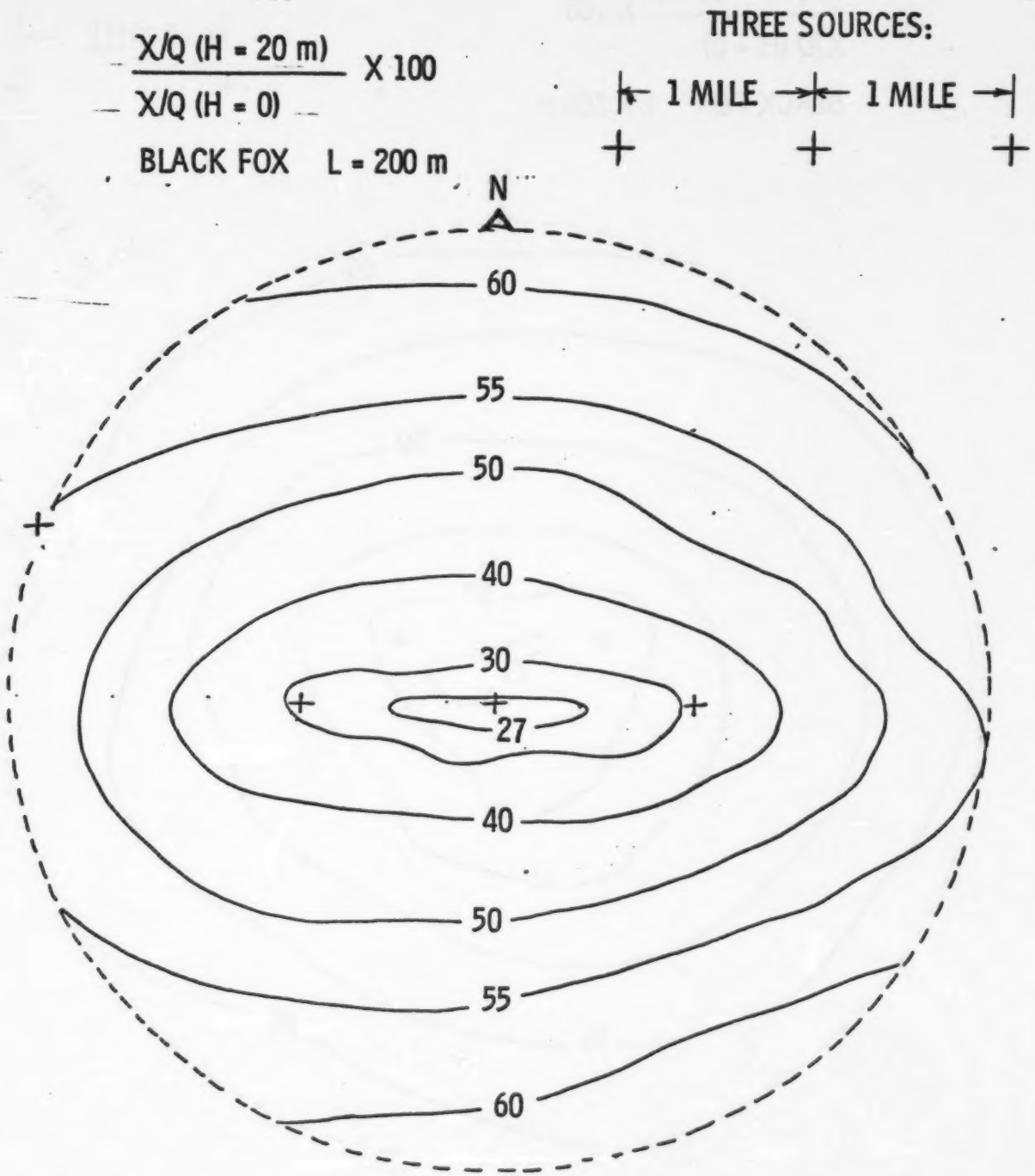


Figure 10c. Areal Distribution of Ratio of Ground Level χ/Q (H=20m) to Ground Level χ/Q (H=0) for Black Fox Meteorology. Mixing Height of 200m. Area Within 2 Miles of Black Fox. Depicted Triple Source Configuration.

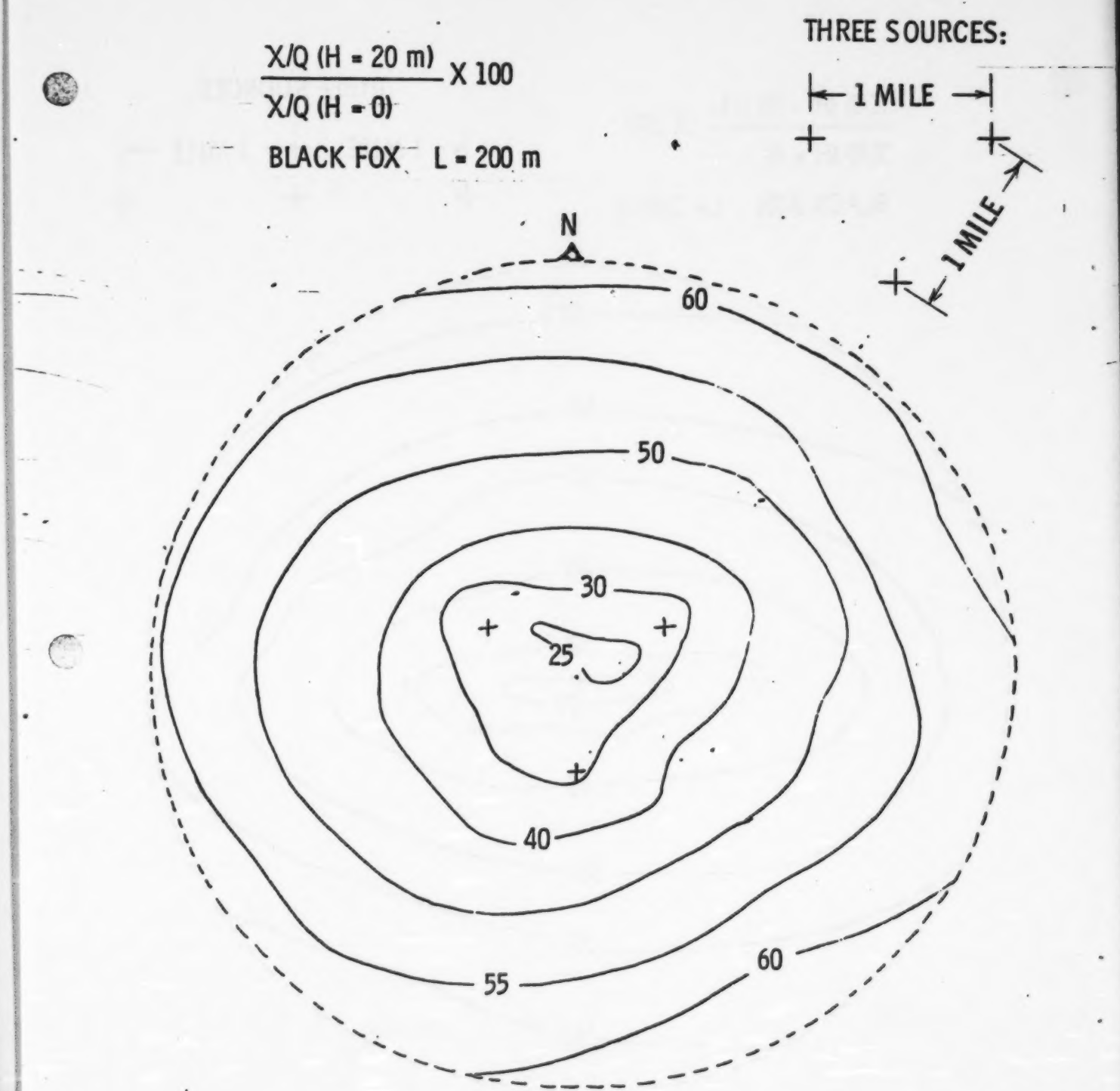


Figure 10d. Areal Distribution of Ratio of Ground Level x/Q (H=20m) to Ground Level x/Q (H=0) for Black Fox Meteorology. Mixing Height of 200m. Area Within 2 Miles of Black Fox. Depicted Triple Source Configuration.

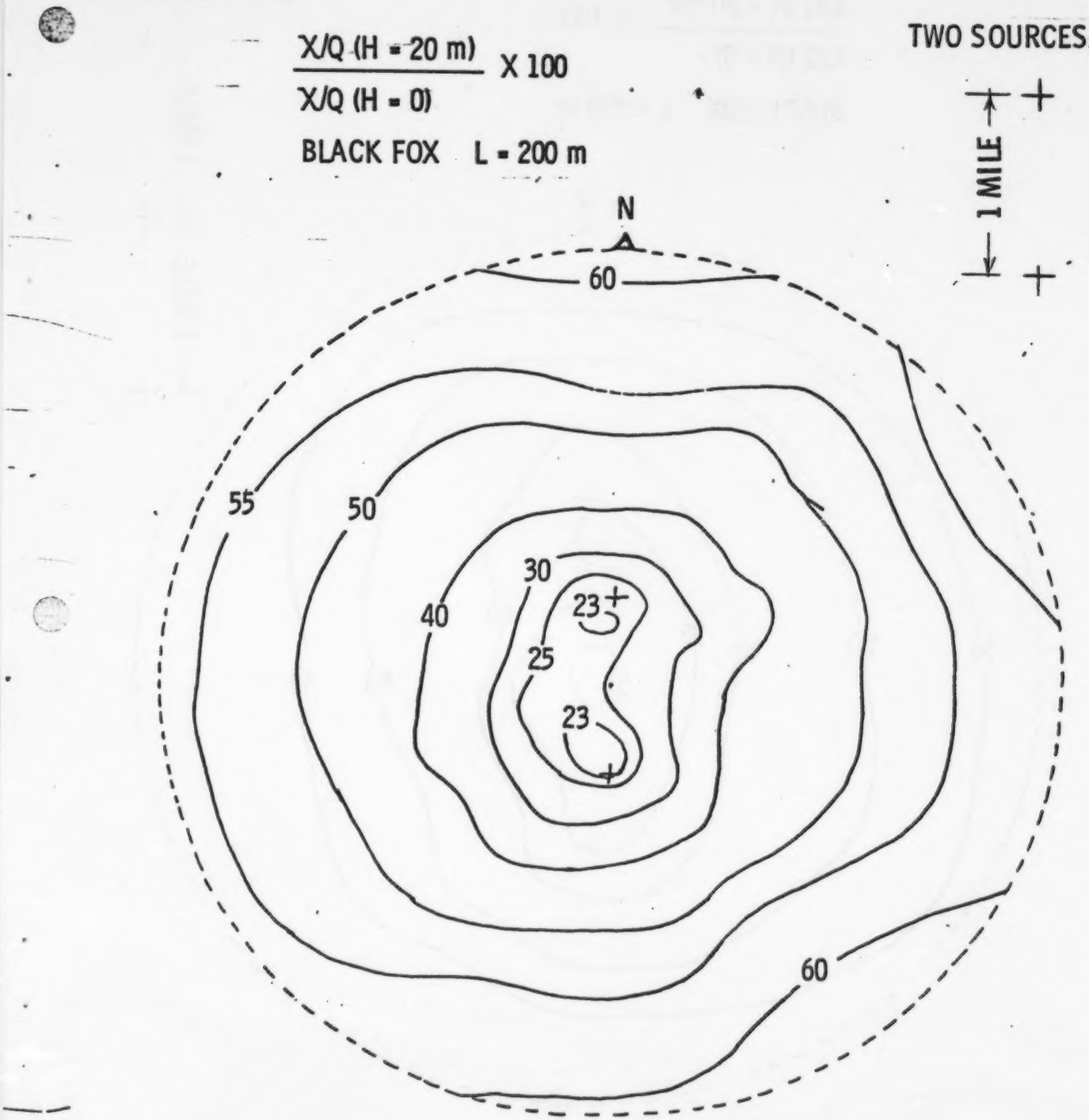


Figure 10e. Areal Distribution of Ratio of Ground Level χ/Q (H=20m) to Ground Level χ/Q (H=0) for Black Fox Meteorology. Mixing Height of 200m. Area Within 2 Miles of Black Fox. Depicted Dual Source Configuration.

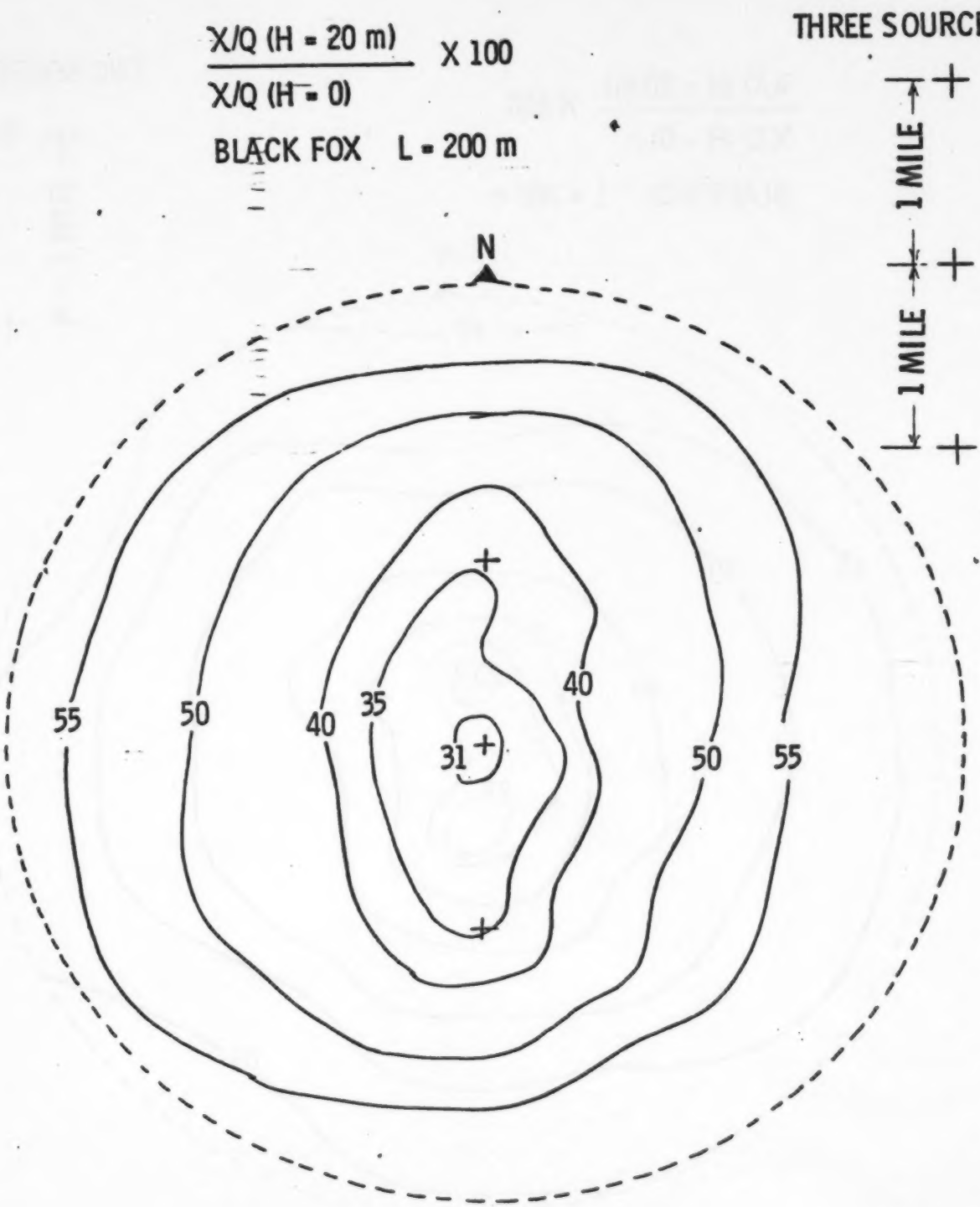


Figure 10f. Areal Distribution of Ratio of Ground Level χ/Q (H=20m) to Ground Level χ/Q (H=0) for Black Fox Meteorology. Mixing Height of 200m. Area Within 2 Miles of Black Fox. Depicted Triple Source Configuration.

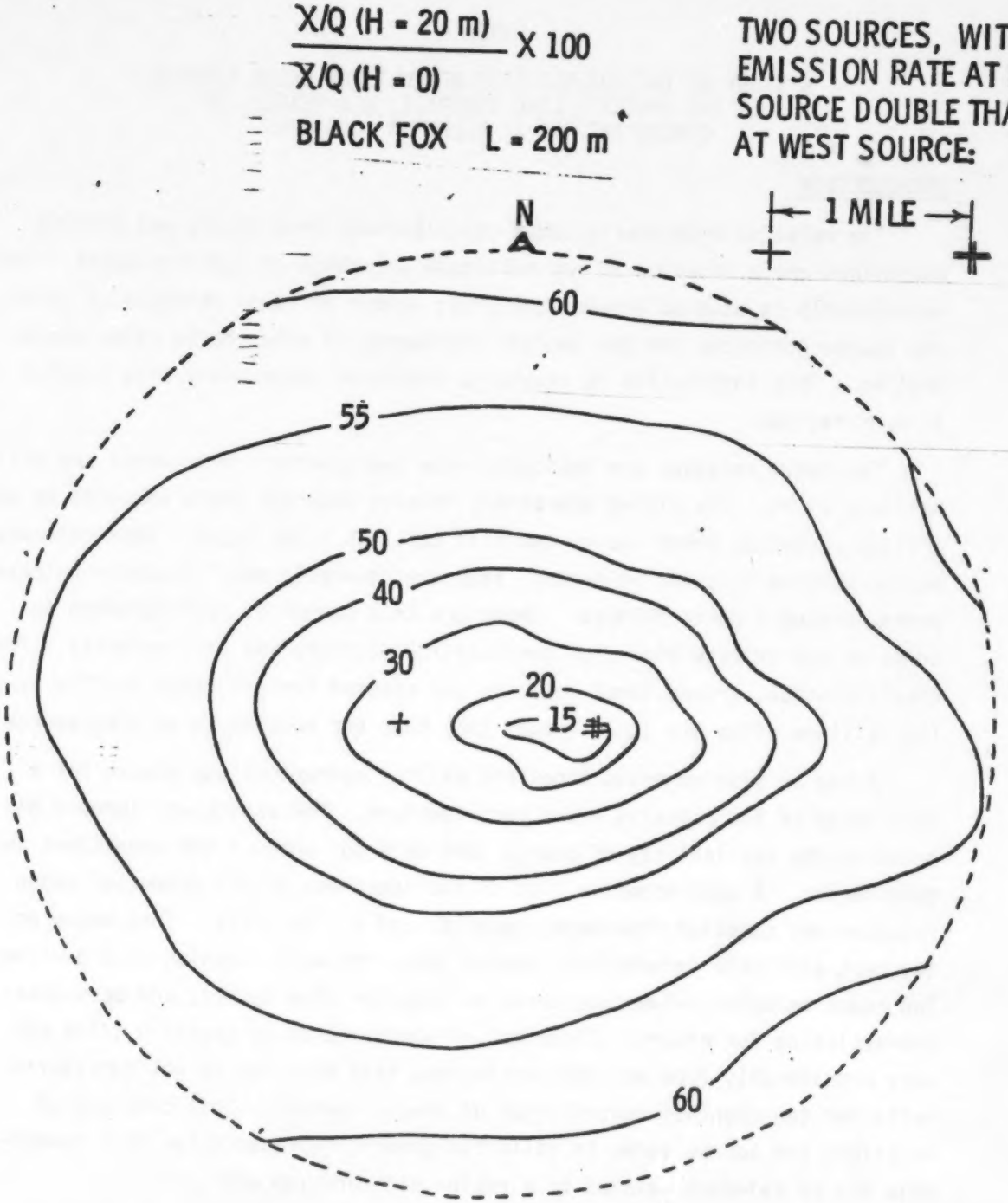


Figure 10g. Areal Distribution of Ratio of Ground Level χ/Q (H=20m) to Ground Level χ/Q (H=0) for Black Fox Meteorology. Mixing Height of 200m. Area Within 2 Miles of Black Fox. Depicted Dual Source Configuration.

TASK 4

A STUDY OF THE CONTRIBUTION OF MULTIPLE RADON SOURCES IN THE AMBROSIA LAKE DISTRICT, NEW MEXICO, TO CONCENTRATIONS AT SELECTED LOCATIONS

INTRODUCTION

The relative atmospheric radon contributions from mining and milling operations are a function of the multitude and range of radon releases. The relationship is studied between the great number of radon releases of various source strengths and the implied increments in atmospheric radon concentration. This information is needed to interpret radon monitoring results in such regions.

The radon releases are typically from two sources: mine vents and mill tailings piles. The mining operations involve numerous vents compared to the milling operation where one or two mill tailings piles occur. Vent releases may be modeled as point releases. They are generally small diameter releases, approximating a point release. These are in a number of configuration in terms of the release direction (vertical/horizontal) and exit velocity. For simplification, ground level releases are assumed for all vents in this study. The tailings piles are large enough that they are modeled as an area source.

A region with numerous mines and milling operations was chosen for a case study of the relative radon contributions. The choice was largely dictated by the availability of source term data for some of the operations in this region. A comprehensive list of the locations of all potential radon releases was compiled from maps, reports, and a site visit. Then based on the best available information, source terms for each location were derived. The radon emission values are based on data for some points, and best-guess extrapolation for others. Since actual source terms at specific sites may vary considerably from our best estimates, this data set is not considered valid for the absolute contribution of single sources. This data set of locations and source terms is valid for generic representation of a reasonable mix of releases related to a region with uranium operations.

The atmospheric dispersion and transport are modeled using a ground level straight-line model. The relative contributions of the various re-

leases are summarized at selected distances downwind of the sources.

DISCUSSION

The radon sources identified for this region included 20 mines with about 100 vent or other release locations and four uranium mill sites with a mill tailings pile at each. The mine releases varied from tens of curies per year to several with about 5000 curies per year, and one with 8000 curies per year. The average release was 1070 curies per year. Table 1 gives a breakdown of the radon emissions for selected ranges of emission for both vents and mill tailings piles. Most of the vent sources can be seen to be near the 1000 curies per year average.

TABLE 1. Frequency of Radon Emission Ranges in the Study Region

Number of Sources	Range of Source Terms (Ci/yr)						
	0 to 100	101 to 1000	1001 to 2000	2001 to 3000	3001 to 4000	4001 to 5000	Greater than 5000
Vents	12	62	21	7	1	3	4
Mill Tailings	0	0	2	0	0	1	1

The estimates of radon emissions from the four mill tailings piles ranged from 1600 to 5000 curies per year with an average value of 3100 curies per year. These emissions are the same order as the individual larger mine vents. The areal extent of the mill tailings piles reduces the initial downwind concentration in comparison with similar release rates from vents.

In terms of magnitude of releases of radon, the largest individual releases are from a few mine vents and the mill tailings piles. The actual orientation and spacing of the releases relative to the winds in a given region will control the specific regional incremental combinations of radon.

A search was conducted to identify appropriate annual meteorological data to assess the long-term radon contributions from these sources. An annual summary obtained from a station near the region was judged inapplicable

to the region. The complexity of flow in the region makes onsite data a necessity. One year of meteorological data taken in a central location in this region was obtained from the Office of Radiation Programs, Environmental Protection Agency, Las Vegas, Nevada. These data are in a ten-minute format and are being reduced to a format appropriate for long-term evaluation of radon incremental air concentrations. Environmental exposures to radon are generally considered in terms of long-term exposure. Short-term concentrations are of interest in terms of occupational exposures and contributions to long-term exposures.

A relative comparison of the possible combinations of contributions to radon air concentrations may be obtained by converting the emissions to air concentrations at selected downwind distances. The range of possible combinations is given by two values--the single release and the combination of sources into single point release. The former is the smallest possible contribution assuming all sources are located such that there is no significant combination of radon plumes. The latter is the case where all sources are at one point. The closest real case is to have all the sources on a line along the wind direction. The real contributions will lie somewhere between these extremes.

The atmospheric increments from radon sources are compared under restrictive atmospheric dispersion conditions [wind speed = 1 m/s and atmospheric stability "F" (Turner, 1969)] to provide relative comparisons of the sources. The range of concentrations at 1 km from one source and 62 sources for the second emission group (101-1000 in Table 1) are 12 to 760 pCi/ℓ. For comparison, the equivalent value for the largest vent release is 190 pCi/ℓ, and for the largest mill tailings pile is 5.8 pCi/ℓ. The values are hourly maximum values under atmospheric conditions that will occur infrequently and not for extended time periods. Actual average exposure values will be orders of magnitude lower reflecting the actual mix of atmospheric conditions.

Table 2 provides a summary of the relative values of the radon source terms in terms of percentages. At greater distances downwind, the release characteristics are lost as a controlling factor in the resultant air concentrations. These values reflect the relative importance on incremental radon

concentrations of the various sources at extended distances downwind. The vents are by far the greatest source, with the multitude of smaller releases contributing as much as the few larger releases. Mill tailings contribute slightly over 9% of the total.

TABLE 2. Relative Source Terms from Uranium Mining and Milling Operations in Percentage of Total

Number of Sources	Range of Source Terms (Ci/yr)						
	0 to 100	101 to 1000	1001 to 2000	2001 to 3000	3001 to 4000	4001 to 5000	Greater than 5000
Vents	0.4%	25.0	23.1	12.9	2.6	10.9	18.6
Mill Tailings	0	0	2.2	0	0	3.2	3.7

The effort to assess the long-term contributions of incremental radon concentrations is to be reported under separate cover. The one year of on-site meteorological data is being used to compute the environmental exposures at selected residential and nonresidential locations in the region.

SUMMARY

Uranium mining and milling operations result in a multitude of radon releases of varying source strength. An analysis is performed of the relative importance of these various radon releases on ambient atmospheric radon values. A uranium mining and milling region is selected to provide case study data on the locations, numbers, and magnitudes of releases. A comprehensive list of radon releases is compiled. Then using the best available information, source terms are generated for each location. Estimates of the atmospheric transport and dispersion of these emissions provide a framework for comparison of the relative contributions of atmospheric radon. The mine vent radon releases contribute a much larger fraction of the incremental atmospheric radon values than tailings piles. Although a few mine vents do have large radon emission rates, the large number of smaller mine releases constitutes a major fraction of total radon emissions.

RECOMMENDATIONS

The source term data have clearly pointed out the importance of the vent releases in total radon incremental concentrations. Future studies should have stronger emphasis on the vent releases. The models should account for actual configuration of vent releases (size, elevation, exit velocity, etc.). The assumption of ground level releases may be resulting in considerable overestimation of short-range concentrations.

Verification of the modeled concentrations should be made using monitoring data and onsite studies of specific dispersion conditions.

The results should be generalized to other regions by comparisons of the vent configuration, numbers, and locations. Should the annual radon concentrations be limiting, similar efforts should be undertaken for other regions.

References

Turner, D. B. 1969. Workbook of Atmospheric Dispersion Estimates, USDHEW, PHS Pub. No. 995-AP-26.

APPENDIX

RADON MEASUREMENT BY CHARCOAL SAMPLING

Radon collection was made using small charcoal canisters containing approximately 75 grams of activated charcoal. The collection system was assembled by mating the charcoal canister with a small lucite cylinder to hold the charcoal aboveground and define the exact area of exhalation. A thin tedlar film served to cover the top surface of the canister, thus allowing external pressure variations to be more closely approximated on the surface being sampled.

At Morton Ranch, these charcoal canisters were exposed for one day. At the mine areas in the Grants, New Mexico region, canisters were exposed for a minimum of four days and a maximum of ten days. Samples from the Wyoming area were analyzed after attainment of radon daughter equilibrium at the Pacific Northwest Laboratory, Richland, Washington, using multidimensional gamma-ray spectrometers. These systems are described in Nuclear Instruments and Methods, Vol. 50, 1967, pages 1 through 10. Briefly, the systems are fabricated from large 11" diameter x 6" thick NaI(Tl) activated crystals which are separated from their phototubes by 4" thick pure NaI lightpipes. Each NaI(Tl) crystal is surrounded by a plastic phosphor or NaI(Tl) anticoincidence annulus. The spectrometer is coupled to a 4096-channel multiparameter analyzer and uses the gamma-ray decay characteristics of each radionuclide for its identification and measurement. The final data analysis is done with a PDP-15 computer program which allows the measurement of radon through its gamma-emitting daughter products. Charcoal canisters from the Grants, New Mexico area were, in general, analyzed at the EPA Montgomery laboratory.

RADIUM, THORIUM, AND POTASSIUM RADIONUCLIDE ANALYSES

A variety of multidimensional gamma-ray spectrometers were used to analyze the particulates in water, air filter particulates, and soils obtained in the Grants, New Mexico and Douglas, Wyoming uranium mining areas. These gamma-ray detection systems utilize pairs of NaI(Tl) activated crystals which are attached to their phototubes by pure NaI lightpipes. Each NaI(Tl)

activated crystal is surrounded by a plastic phosphor or NaI(Tl) anticoincidence shield. Each spectrometer is coupled to a 4096-channel multiparameter analyzer and uses the gamma-ray decay characteristics of each radionuclide for its identification and measurement. This is accomplished by viewing the sample with the two NaI(Tl) detectors operating in coincidence and cancelling those events not totally absorbed in the principal detectors by the anticoincidence annulus. Coincidence counts are stored in an energy-energy plane of the memory according to their photon energies, while single gamma rays which interact with either crystal are stored in a normal manner on the X or Y axis. The axis and energy-energy plane backgrounds, backscatter effects, Compton response, and efficiency parameters are stored in a PDP-35 computer memory and used to analyze the collected gamma spectrometer data. In this fashion, ^{228}Th was measured in all samples using the ^{208}Tl gamma rays. Radium-226 was analyzed using the photons from ^{214}Bi and ^{40}K was measured with its single 1.46 MeV photon. Counting was delayed for several weeks after sealing the samples in aluminum cans to permit a near-equilibrium build-up of radon, and hence ^{214}Bi, from which the ^{226}Ra was inferred.

TRACE ELEMENT ANALYSIS IN AQUEOUS SAMPLES

Fifteen trace elements were analyzed in the water samples by two techniques. Plasma emission spectrometry was used for the elements molybdenum, copper, beryllium, aluminum, nickel, cobalt, cadmium, manganese, barium, vanadium, zinc, and iron. Sample pretreatment consisted simply of addition of a small amount of lithium ionization buffer. Samples were run directly against a single standard with double distilled water plus lithium used as the instrument blank. The instrument used was an SMI Echelle spectrometer with a Spectrospan III D.C. argon plasma excitation source. Analysis was carried out in sequential single element mode. A graphite furnace atomic absorption procedure was used for lead, arsenic, and selenium to obtain adequate sensitivity for these elements. Lead samples were run directly as received. Samples for arsenic and selenium were made 0.05% in nickel to minimize losses during pyrolysis. The instrument used was an ILA 257 atomic absorption spectrometer with a Model 555A graphite furnace. Samples

were arpirated onto a cylindrical pyrolytic graphite cuvette using an ILA FASTAC autosampler in semi-automatic mode. Standardization was by freshly prepared dilution of stock AA standards. It should be noted that of the trace elements originally requested, no data are reported for antimony or mercury. High sensitivity analysis of antimony is possible by GFAA; however, since no antimony lamp was available, it could not be done within the required time scale. Mercury analysis requires special sampling techniques which were not utilized on these samples. We therefore felt that any mercury data obtained on these samples would not be meaning and did not attempt to analyze for mercury.

ELEMENTAL ANALYSIS OF SOILS BY X-RAY FLUORESCENCE (XRF)

The granular geological grab samples were homogenized, coned, and quartered to select a representative aliquot of about 4 grams. This aliquot was then reduced to near 100 mesh by alumina mortar and pestle. Samples of approximately 400 mg were then prepared for X-ray fluorescence analysis by forming wafters of 7.9 cm^2 at 3700 kg/cm^2 pressure. These wafers were sequentially fluoresced with three excitation sources to provide a broad range of elemental analyses and maximum sensitivities. The sources used were a zirconium secondary source from a tungsten tube excited at 50 kV and 14 mA; a silver secondary source from a tungsten tube excited at 45 kV and 25 mA; and a ^{241}Am isotopic source. The zirconium excitation lasted 30 minutes, the silver excitation 45 minutes, and the ^{241}Am excitation 60 minutes.

The fluorescence systems employed were KEVEX Model 810 tube-excited system and a radioisotope-excited laboratory system. X-ray spectra were collected with an 80 mm^2 x 5 mm Si(Li) diode and accumulated by computer-based multichannel analyzers (Models 4410 and 4420, Nuclear Data, Inc.). Reduction of the multichannel-analyzer data to elemental concnetrations was accomplished by a PDP 11/35 computer using FORTRAN programs SAP and SAP3 (Nielson, 1977).* Analysis of particulate matter in the aqueous samples was accomplished by fluorescing the 0.4 μm filters directly and analyzing the resulting data in terms of the total volume of liquid passed through the filter.

*Nielson (1977) SAP3: A Computer Program for X-ray Fluorescence Data Reduction, BNW-2193, Pacific Northwest Laboratory, Richland, Washington.

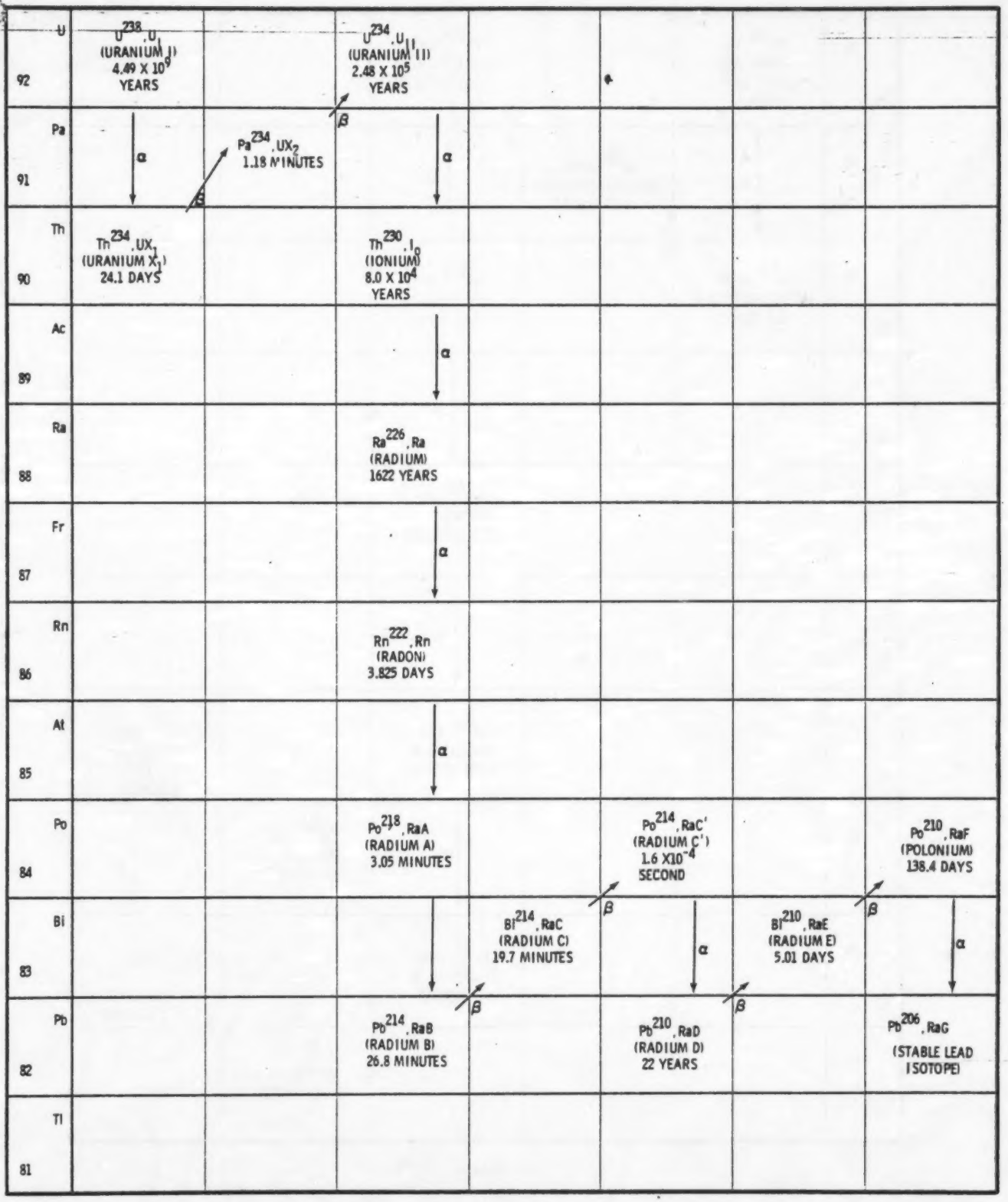
ELEMENTAL ANALYSIS OF SOILS BY INSTRUMENTAL NEUTRON ACTIVATION ANALYSIS (INAA)

Soil samples (0.5-1.0 gm each) were analyzed for 31 major, minor, and trace elements by well developed sequential INAA procedures using a high efficiency Ge(Li) detector (25%, FWHM 1.8 KeV for 1332 KeV γ of ^{60}Co) and a 2048-channel analyzer. The INAA procedure consisted of one short and one long irradiation and two counting intervals in each irradiation. In short irradiations, the samples were individually irradiated in a pneumatic facility at a flux of $\sim 5 \times 10^{11}$ n/cm^2/sec (100 KW) for 2 minutes each, delayed for 10 minutes and counted for 5 minutes each on the Ge(Li) system. The elements titanium, vanadium, magnesium, and aluminum were measured in this counting. After a delay of 2-4 hours, the samples were again counted for 10 minutes each to measure manganese and sodium. The only mjaor interference is from the reaction Al(n,p) Mg which is estimated as 1% Al = 0.28% Mg. This correction is significant and the reported Mg values contain large uncertainties.

Following the short irradiation, the samples were irradiated at a higher flux of $\sim 5 \times 10^{12}$ n/cm^2/sec (1 MW) for about 4 hours. After a decay of 3-4 days, the samples were counted for 30-60 minutes each on the Ge(Li) system. The elements sodium, potassium, calcium, arsenic, bromine, antimony, barium, lanthanium, neodymium, samarium, ytterbium, lutetium, and uranium were measured in this counting. After another delay of 3-4 weeks, the samples were again counted for 60-200 minutes each to measure scandium, chromium, iron, cobalt, rubidium, zirconium, cesium, cerium, europium, terbium, hafnium, tantalum, and thorium. The only significant interference is from the uranium contributions in the samarium and zirconium determinations, which was corrected. The usual estimated errors based on counting statistics are: ±0.5-5% for sodium, aluminum, scandium, manganese, iron, cobalt, lanthanium, samarium, and ytterbium; ±5-10% for potassium, calcium, titanium, vanadium, chromium, arsenic, bromine, rubidium, antimony, cesium, barium, europium, hafnium, tantalum, thorium, and uranium; ±10-20% for magnesium, zirconium, cerium, neodymium, terbium, and lutetium.

DECAY SCHEMES FOR ^{238}U AND ^{232}Th

The following decay schemes illustrate the inter-relationships of the various radionuclides discussed in this report and also indicate half-lifes of many of the species which were measured.



THE URANIUM SERIES

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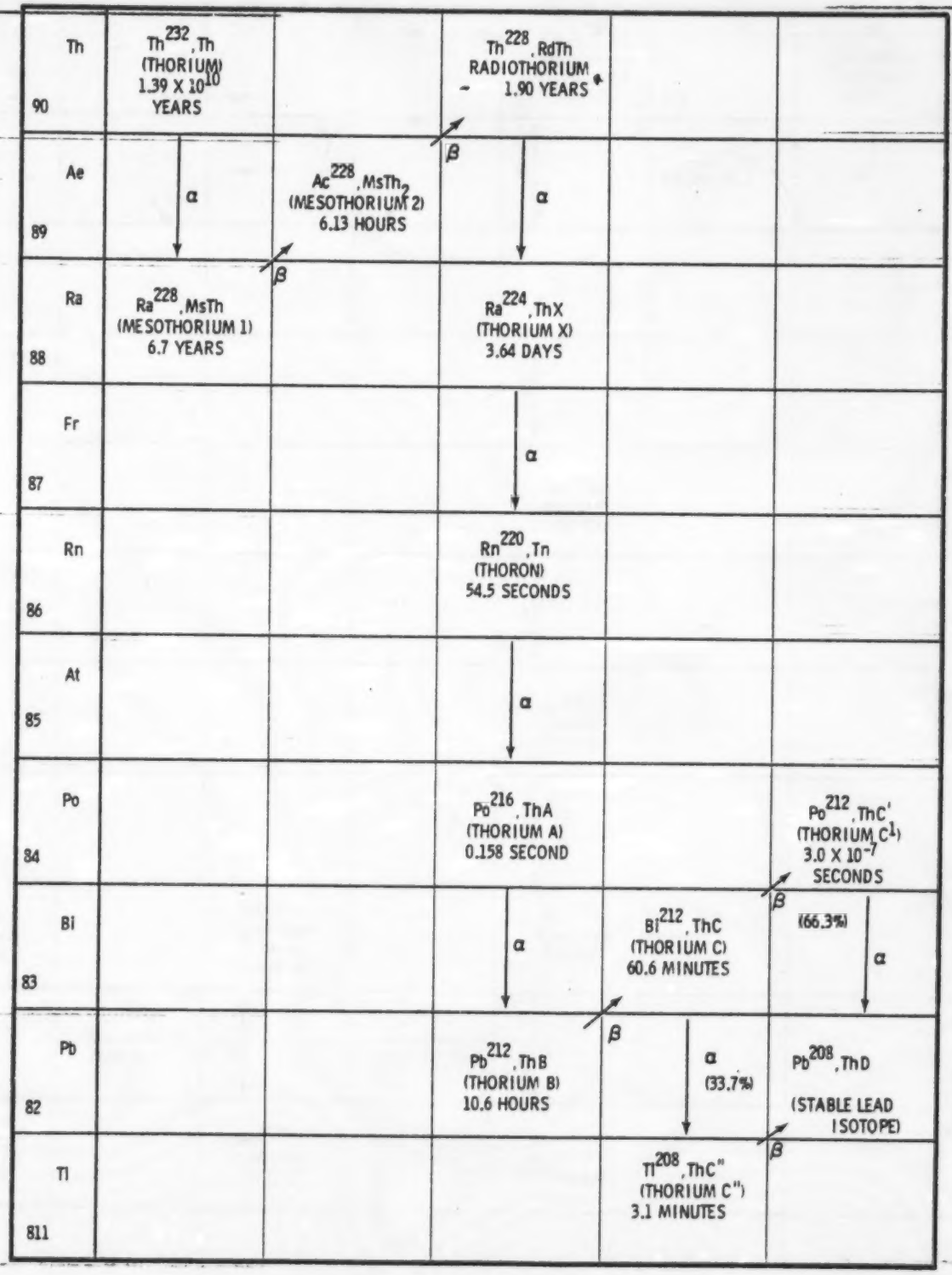

THE THORIUM SERIES